

Best Available Copy

4



Europäisches Patentamt

⑩

European Patent Office

Office européen des brevets

⑪ Publication number:

0 049 982

B1

⑫

EUROPEAN PATENT SPECIFICATION

⑯ Date of publication of patent specification: 21.05.86

⑮ Int. Cl. 4: D 01 F 8/00, D 01 D 5/30

⑰ Application number: 81304583.8

⑯ Date of filing: 02.10.81

④ Novel elastic fibers, an assembly thereof, and process for production of said assembly.

⑩ Priority: 02.10.80 JP 136696/80
02.10.80 JP 136697/80
16.10.80 JP 143685/80⑩ Proprietor: TEIJIN LIMITED
11 Minami Honmachi 1-chome Higashi-ku
Osaka-shi Osaka-fu (JP)⑩ Date of publication of application:
21.04.82 Bulletin 82/16⑩ Inventor: Emi, Shingo
98-16 Minamiwakuni-cho 2-chome
Iwakuni-shi Yamaguchi-ken (JP)
Inventor: Norota, Susumu
1-21 Yamate-cho 3-chome
Iwakuni-shi Yamaguchi-ken (JP)
Inventor: Kiriyama, Tsutomu
1-3 Yamate-cho 3-chome
Iwakuni-shi Yamaguchi-ken (JP)
Inventor: Segawa, Yasuhiko
28-4-208 Ozu-cho 1-chome
Iwakuni-shi Yamaguchi-ken (JP)
Inventor: Imoto, Tadasi
10-18 Katsura-machi 2-chome
Iwakuni-shi Yamaguchi-ken (JP)⑩ Publication of the grant of the patent:
21.05.86 Bulletin 86/21⑩ Designated Contracting States:
DE FR GB⑩ References cited:
EP-A-0 017 423
FR-A-2 142 419
GB-A-1 175 192
GB-A-1 558 592⑩ Representative: Woods, Geoffrey Corlett et al
J.A. KEMP & CO, 14 South Square Gray's Inn
London WC1R 5EU (GB)

EP 0 049 982 B1

The file contains technical information
submitted after the application was filed and
not included in this specification

Note: Within nine months from the publication of the mention of the grant of the European patent, any person may give notice to the European Patent Office of opposition to the European patent granted. Notice of opposition shall be filed in a written reasoned statement. It shall not be deemed to have been filed until the opposition fee has been paid. (Art. 99(1) European patent convention).

Description

This invention relates to novel elastic fibers, an assembly of said fibers, and a process for producing said assembly. More specifically, it relates to novel elastic fibers having improved elastic properties and composed of at least two thermoplastic polymers, an assembly of said fibers, and a process for producing said assembly.

"Spandex" filaments made from a polyurethane elastomer and elastic filaments made from a polyester elastomer have been known as fibers having excellent elastic recovery. Elastic fibers which develop elasticity by an entropy mechanism, such as fibers of polyurethane elastomers or polyester elastomers, have the defect that they have a very low initial modulus of elasticity and a rubber-like feel, and tend to block, and when used at low temperatures, their elastic recovery and elongation abruptly decrease. Furthermore, because of their blocking tendency, these entropically elastic fibers are not spinnable on a spinning frame.

On the other hand, polypivalolactone fibers (see U.S. Patents Nos. 3,299,171 and 3,776,997), polybutene-1 fibers (see U.S. Patents Nos. 3,686,385 and 3,840,510), poly(ϵ -capramide) fibers (see U.S. Patent No. 3,513,110), polyoxymethylene fibers (see U.S. Patent No. 3,551,363), and polypropylene fibers (U.S. Patents Nos. 3,256,258; 3,323,190; 3,485,906; 3,377,415; and 3,432,590) are known as elastic fibers having a high initial modulus of elasticity. Productivity of these fibers, however, is very low because they are produced by a method which comprises melt-spinning a polymer, and subjecting the resulting filaments, either as such or after being drawn at a low ratio, to dry or wet heat treatment or pressurizing treatment for a long period of time.

These elastic fibers develop elasticity by a crystal elasticity mechanism as contrasted to the aforesaid entropy elasticity mechanism, and have a very high initial modulus of elasticity. Accordingly, these fibers are called "hard elastic fibers" [see J. Macromol. Sci. Phys. B8 (1-2), 157 (1973) and Colloid & Polymer Sci. 253, 824-839 (1975)]. Although these crystalline elastic fibers are excellent in that they retain elastic properties at low temperatures in the same way as at room temperature unlike the aforesaid entropically elastic fibers, their excessively high initial modulus of elasticity and low elongation make them unsuitable for apparel applications.

EP-A-17423 (published 15 October 1980) describes filaments composed of at least one synthetic polymer, the filament having an irregular variation in size along its longitudinal direction and a coefficient of intrafilament cross-sectional area variation [CV(F)] of 0.05 to 1.0. Mixtures of synthetic thermoplastic polymers may be used in making such filaments and mention is made of polymers capable of forming a row structure and of polymers capable of forming lamellar crystals. There is, however, no disclosure or suggestion of elastic filaments having a row structure.

To the best knowledge of the inventors, elastic fibers which stand between the entropically elastic fibers and the hard elastic fibers and can find extensive uses, especially those having excellent elastic recovery, a moderate initial modulus of elasticity and a moderate elongation, have not been provided to date.

The invention seeks to provide novel elastic fibers composed of at least two thermoplastic polymers, having a moderate initial modulus of elasticity and having excellent elastic recovery.

According to the invention there is provided a fiber wherein

- (a) said fiber is composed of at least two thermoplastic polymers,
- (b) at least one of said thermoplastic polymers is capable of forming a row structure and
- (c) at least one other of said thermoplastic polymers is capable of forming lamellar crystals,
- 45 characterised in that
- (d) said fiber is elastic, has a row structure, an elastic recovery ratio (ER) after 50% stretch of at least 70% and an elongation (EL) of from 200 to 700%.

The invention also provides an elastic fibrous assembly of a plurality of said elastic fibers and a process for producing such an assembly.

Elastic fibers composed of at least two thermoplastic polymers have not been known heretofore as stated above. It is known that elastic fibers composed of a thermoplastic polymer having crystallinity capable of forming a row structure, such as polypropylene or polyoxymethylene, have a row structure in which lamellar crystals generated by folding of polymer molecule chains are stacked in the direction of C axis (see J. Macromol. Sci.-Phys., B1, 41, 1967 and J. Macromol. Sci.-Phys., B5, 591, 1971). These elastic fibers having such a row structure are believed to be crystalline elastic fibers which develop elasticity by a mechanism wherein lamellae or lamellar bundles are opened by the effect of interconnecting tie molecules present between the lamellae or lamellar bundles, as Clark et al. show by a schematic diagram [E. S. Clark, C. A. Garber, Int. J. Polymeric Materials, 1, 31 (1971)]. For this reason, mixing of a polymer giving such crystalline elastic fibers with another polymer would be considered to hamper the formation of lamellar crystals and consequently a row structure and to be undesirable for production of elastic fibers.

Surprisingly, it has now been found in accordance with this invention that by using a combination of a thermoplastic polymer capable of forming a row structure and another thermoplastic polymer capable of forming lamellar crystals, the formation of a row structure is not hampered but is rather promoted to give elastic fibers having improved elastic properties.

0 049-982

The process for producing the elastic fibers of this invention is first described in detail with reference to the accompanying drawings.

Brief description of the accompanying drawings

5 Figure 1-a is a schematic view of a wire mesh which is one example of a mesh spinneret used in the production of the elastic fibers of the invention;
Figure 1-b is a partial longitudinal sectional view of Figure 1-a;
Figure 2 is a rough sketch of one example of an apparatus suitable for the production of the elastic fibers of the invention;
10 Figure 3 is a generalized schematic view of any arbitrary longitudinal section of a mesh spinneret used in the process of this invention;
Figure 4 is a schematic longitudinal sectional view of one example of a die used in the production of the elastic fibers of the invention;
Figure 5 is a photograph of a section of a molten polymer in Example 29 in which polymer (b) and
15 polymer (c) form relatively large blocks when they reached the mesh spinneret;
Figure 6 is a photograph showing a section of one example of conjugate elastic fibers in which polymer (b) and polymer (c) respectively form relatively large polymer phases in the cross section of the fibers;
Figure 7 is a photograph of a section of elastic fibers in which polymer (b) and polymer (c) are finely dispersed in the cross section of the fibers;
20 Figures 8-a and respectively 8-b are broad angle X-ray photographs of the elastic fibers obtained in Comparative Example 1 and Example 12; and
Figures 9-a and 9-b are electron microphotographs of the surface of the elastic fibers obtained in Example 24, which respectively show the crystals of the row structure.

25 Manufacturing apparatus and process

The elastic fibers of this invention can be produced by using any spinning apparatuses for melt spinning. For example, the elastic fibers can be produced by a known method which comprises melt-extruding at least two thermoplastic polymers, either as a mixture or as conjugate fibers, from
30 uniform regularly-shaped orifices provided at certain intervals in a spinneret. According to this invention, the elastic fibers can be produced by heat-treating the as-spun fibers. The elastic fibers of the invention provided by the above known spinning and the heat treatment have a uniform and fixed cross sectional shape conforming to the geometric configuration of the orifices.

In the production of the elastic fibers of the invention by the above process, it is desirable to perform
35 the spinning at a take-up speed of 10 to 200 m/min and a draft ratio of 10 to 100 while quenching the filaments as much as possible, although these conditions may vary depending upon the types of the polymers used.

According to the above process, it is relatively difficult, in comparison with a process to be described, to produce the elastic fibers of the invention having the desired improved elastic properties by
40 heat-treatment of the resulting as-spun fibers. This is presumably because according to the aforesaid spinning method, the quenching is relatively difficult, and a longer period of time is needed to heat-treat the resulting as-spun fibres than as-spun fibers obtained by the process to be described below using a mesh spinneret when it is desired to impart the desired elastic properties. In addition, the aforesaid process apparently has low productivity. Accordingly, the aforesaid process is not so recommendable for
45 production of the elastic fibers of the invention.

According to the process of this invention an elastic fibrous assembly of a plurality of the elastic fibers is produced by extruding a combined polymer melt of at least two thermoplastic polymers through a mesh spinneret, the combined polymer melt comprising at least 10% by weight of at least one thermoplastic polymer (b) which is capable of forming a row structure and not more than 90% by weight of at least one other thermoplastic polymer (c) which is capable of forming lamellar crystals, said spinneret including many closely arranged small openings and having an opening ratio (α), represented by the following formula, of at least 10%,

$$55 \quad \alpha = \frac{V_a - V_i}{V_a} \times 100$$

in which V_a represents the total apparent volume of the spinneret which is taken within a unit area of its mesh portion, and V_i represents the total volume of partitioning members defining the small openings which is taken within a unit area of the mesh portion of the spinneret; said extrusion being carried out while generating Joule heat in the partitioning members of the spinneret and cooling the vicinity of the extrusion surface of the spinneret by supplying a cooling fluid, whereby the melt is converted into fine streams by the partitioning members; taking up and solidifying the fine streams without drawing them to provide as-spun fibers; and heat-treating the resulting as-spun fibers.

65 The spinneret used in this process includes many closely arranged small openings defined by the

0 049 982

opening ratio (α). In the above formula defining the opening ratio, the mesh portion of the spinneret denotes that portion of the spinneret which is mesh-like.

So long as the spinneret used in this invention includes many closely arranged small openings defined by the above opening ratio, there is no particular restriction on the shape of the small openings, and the shapes of the partitioning members defining the small openings. Accordingly, the mesh spinneret used in this invention may have a circular, elliptical, triangular, tetragonal, or polygonal shape, or the partitioning members defining the small openings may have depressions and elevations.

Figure 1-a of the accompanying drawings illustrate a typical example of the mesh spinneret used in this invention. The illustrated mesh spinneret is a plain weave wire mesh, and its cross section is shown in Figure 1-b. In the plain weave wire mesh illustrated in the drawings, a small opening is of a tetragonal shape and a partitioning member defining this small opening has a depression through which a combined polymer melt extruded from the small opening moves toward and away from a combined polymer melt extruded from an adjacent small opening.

The mesh spinneret used in this invention may also be an etched porous plate made by providing many small openings on a thin metallic plate by an elaborate etching technique. The etched porous plate has many small openings of a trilobal shape and a partitioning member present between adjacent small openings has a depression.

The mesh spinneret used in this invention may also be a twill weave wire mesh, or a thin sintered body obtained by sintering many minute metallic balls so as to form many small openings. A part of the mesh spinneret used in this invention is disclosed in EP—A—17423.

The mesh spinneret used in this invention may be used singly or as a laminated assembly.

The spinneret used in this invention is preferably a mesh spinneret having many small openings defined by partitioning members of small width having elevations and depressions on its polymer extruding surface, said small openings being such that the combined polymer melt extruded through one small opening of the spinneret can move toward and away from the combined polymer melt extruded from another small opening adjacent to said one opening or *vice versa* through depressions of the partitioning members.

In the above formula defining the opening ratio (α) of the mesh spinneret used in this invention, V_a is the total apparent volume of the spinneret which is taken within a unit area of its mesh portion and V_t is the total volume of partitioning members defining the small openings which is taken within a unit area of the mesh portion of the spinneret.

Again, as is seen from Figures 1-a and 1-b, the total apparent volume (V_a) is defined as a volume formed by two phantom planes of a unit area (1 cm^2) which contact the front and back surfaces of the spinneret.

In practice, the V_a value of a certain mesh spinneret can be simply determined by measuring the thickness of the spinneret by means of a dial gauge having a contact surface of 1 cm^2 in area.

The V_t value of a certain mesh spinneret can be determined by cutting it to a predetermined area, and for example, submerging it in a liquid, and measuring the resulting volume increase. V_t is a value obtained by converting the increased volume for each cm^2 of the spinneret.

Since the opening ratio (α) is expressed by the following formula

$$\alpha = \frac{V_a - V_t}{V_a} \times 100$$

It will be understood that if a 1 cm^2 area of the spinneret is used as a standard in determined V_a and V_t , the value showing V_a is the value representing the thickness of the mesh spinneret as illustrated in Figure 1-b.

The mesh spinneret used in this invention has preferably an opening ratio (α) of 20% to 90%.

Furthermore, the mesh spinneret used in this invention preferably has at least 5, more preferably 10 to 10,000, especially preferably 100 to 1,000, small openings per cm^2 .

Furthermore, the mesh spinneret used in this invention has a thickness of preferably not more than 10 mm, more preferably 0.1 to 5 mm, especially preferably 0.2 to 2 mm.

Advantageously, there is used in accordance with this invention a spinneret having the aforesaid structure in which the average distance (\bar{P}) between extrusion openings for the polymer melt on the surface of its fiber-forming area is in the range of 0.03 to 4 mm. Especially advantageously, there is used a spinneret having an extrusion surface with fine elevations and depressions and numerous small openings for polymer which have

- (1) an average distance (\bar{P}) between small openings of 0.03 to 4 mm,
- (2) an average hill height (\bar{h}) of 0.01 to 3.0 mm,

(3) an average hill width (\bar{d}) of 0.02 to 1.5 mm, and

- (4) a ratio of the average hill height (\bar{h}) to the average hill width (\bar{d}), [$(\bar{h})/(\bar{d})$], of from 0.3 to 5.0.

The fiber-forming area, average distance (\bar{P}) between small opening, average hill height (\bar{h}), average hill width (\bar{d}) and small openings as referred to above are defined below.

The average distance (\bar{P}) between small openings, average hill height (\bar{h}), average hill width (\bar{d}), etc. defined in this invention are determined on the basis of the concept of geometrical probability theory.

Where the shape of the surface of the fiber-forming area is geometrically evidence, they can be calculated mathematically by the definitions and techniques of integral geometry.

For example, with regard to the fiber-forming area of a spinneret in which sintered ball-like objects with a radius of r are most closely packed, the following values are obtained theoretically.

5

$$\bar{P} = \sqrt{3} r, \quad \bar{h} = \frac{\pi}{4} r, \quad \bar{d} = \frac{\pi}{2} r.$$

10 Thus, these parameters can be theoretically determined in a spinneret whose surface is composed of an aggregation of microscopic uniform geometrically-shaped segments. Where the spinneret has a microscopically non-uniform surface shape, \bar{p} , \bar{h} , and \bar{d} can be determined by cutting the spinneret along some perpendicular sections, or taking the profile of the surface of the spinneret by an easily cuttable material and cutting the material in the same manner, and actually measuring the distances between small 15 openings, hill heights, and hill widths. In measurement, an original point is set at the center of the fiber-forming area, and six sections are taken around the original point at every 30° and measured. From this, approximate values of \bar{p} , \bar{h} , and \bar{d} can be determined. For practical purposes, this technique is sufficient.

10 The fiber-forming area, as used in this invention, denotes that area of a spinneret in which a fiber 20 bundle having a substantially uniform density is formed. The spinneret is, for example, the one shown at 7 in Figure 2 for preparing a fiber bundle by extruding a molten polymer.

The small opening in the spinneret denotes the first visible minute flow path among polymer extruding and flowing paths of a spinneret, which can be detected when the fiber-forming area of the spinneret is cut by a plane perpendicular to its levelled surface (microscopically smooth phantom surface taken by levelling 25 the surface with fine elevations and depressions) (the cut section thus obtained will be referred to hereinbelow simply as the cut section of the fiber-forming area), and the cut section is viewed from the extruding side of the surface of the fiber-forming area.

Figure 3 shows a schematic enlarged view of an arbitrarily selected cut section of the general fiber-forming area in this invention. In Figure 3, A_i and A_{i+1} represent the small openings. The distance 30 between the center lines of adjoining small openings A_i and A_{i+1} is referred to as the distance P_i between the small openings. The average of P_i values in all cut sections is defined as the average distance \bar{p} between small openings.

That portion of a cut section located on the right side of, and adjacent to, a given extrusion A_i in a given 35 cut section which lies on the extruding side of the surface of the fiber-forming area from the A_i portion is termed hill H_i annexed to A_i . The distance h_i from the peak of hill H_i to the levelled surface of A_i is referred to as the height of hill H_i . The average of h_i values in all cut sections is defined as the average hill height \bar{h} .

The width of the hill H_i interposed between the small openings A_i and A_{i+1} , which is parallel to the levelled surface of the spinneret H_i is referred to as hill width d_i . The average of d_i values in all cut sections is defined as average hill width \bar{d} .

40 In accordance with the above definitions, the spinneret in accordance with this invention is advantageously such that its polymer molding area, i.e. fiber-forming area, has a surface with fine elevations and depressions and numerous small openings which meet the following requirements.

- (1) The average distance (\bar{p}) between small openings is in the range of 0.03 to 4 mm, preferably 0.03 to 1.5 mm, especially preferably 0.06 to 1.0 mm.
- 45 (2) The average hill height (\bar{h}) is in the range of 0.01 to 3.0 mm, preferably 0.02 to 1.0 mm.
- (3) The average hill width (\bar{d}) is in the range of 0.02 to 1.5 mm, preferably 0.04 to 1.0 mm.
- (4) The ratio of the average hill height (\bar{h}) to the average hill width (\bar{d}), \bar{h}/\bar{d} , is in the range of from 0.3 to 5.0, preferably from 0.4 to 3.0.

More advantageously, in addition to prescribing the values of \bar{p} , \bar{h} , \bar{d} and \bar{h}/\bar{d} within the aforesaid 50 ranges (1) to (4), the structure of the spinneret surface is prescribed so that the value $(\bar{p}-\bar{d})/\bar{p}$ is in the range from 0.02 to 0.8, preferably from 0.05 to 0.7. The value $(\bar{p}-\bar{d})/\bar{p}$, represents the ratio of the area of a small opening within the fiber-forming area.

A characteristic of the process of this invention is that the extrusion of molten thermoplastic polymers is carried out while generating Joule heat in the partitioning members of the mesh portion and cooling the 55 vicinity of the extrusion surface of the spinneret with a cooling fluid.

Accordingly, the partitioning members of the spinneret used in this invention are composed of a conductor material. Examples of the material are metallic elements such as platinum, gold, silver, copper, titanium, vanadium, tungsten, iridium, molybdenum, palladium, iron, nickel, chromium, cobalt, lead, zinc, bismuth, tin and aluminum; alloys such as stainless steel, nichrome, tantalum alloy, brass, phosphor bronze, and Duralmine; and non-metallic conductors such as graphite.

In order to generate Joule heat in the partitioning members of the spinneret, an electric current is directly passed through the spinneret as illustrated in Figure 2.

Joule heat may be generated in the partitioning members of the spinneret by directly passing an electric current through the spinneret as illustrated in Figure 2, or passing an electric current through a coil 65 provided in the inside die of the spinneret to generate an eddy current. The current to be passed may be a

0 049 982

direct current or alternate current in the case of direct supply, but in the case of generating the eddy current, it is an alternate current. According to the process of this invention, it is advantageous to supply a current directly to the spinneret because this permits simplification of the structure of the spinning apparatus.

Usually, a current of 0.1 to several hundred amperes is directly passed through the spinneret, or an electric field of 0.1 to several tens of volts/cm is applied to generate an eddy current. Thus, preferably an energy in an amount of 0.5 to 5,000 watts per cm² of the spinneret is imparted.

According to the process of this invention in which Joule heat is generated from the partitioning members defining the small openings of the spinneret, heat is instantaneously supplied to the thermoplastic polymers at least during its passage through the small openings in contrast to a process in which no heat is generated at the spinneret. As a result, the viscosity and temperature of the polymer melt at the extrusion surface of the spinneret can be controlled to suitable ranges so that the polymer melt can be smoothly separated from the extrusion surface and converted into fine streams.

Since according to the spinneret used in this invention, the polymer melt can be converted to fine streams while supplying a cooling fluid, such as air, to the extrusion surface of the spinneret or its vicinity, the solidification length can be shortened, and the polymer melt can be continuously converted into fine streams stably.

Thus, according to the process of this invention, the solidification length can be shortened, and the temperature of the fine streams can be reduced abruptly from a high temperature.

Accordingly, the process of this invention has the advantage that by short heat-treatment, it can provide as-spun fibers which have crystals grown so that the C axis of the crystals of the row structure can easily extend in the axial direction of the fibers, and which can give the elastic fibers of the invention in which the C axis of the crystals of the row structure extends substantially in the axial direction of the fibers (for example, as-spun fibers produced under optimum spinning conditions give the elastic fibers of the invention having improved elastic properties by heat-treatment for a time period of as short as not more than 1 second).

As can be understood from the above description, the objects and advantages of the invention stated hereinabove can be advantageously achieved by the present invention.

In the process of this invention, the amount of the polymer melt extruded can be adjusted to 0.1 to 20 g/min per cm² of the mesh spinneret.

It is indeed surprising that according to the process of this invention, fine streams of the polymer melt can be more stably spun by turning the extruding surface of the spinneret upwardly so that the normal vector of the extrusion surface is reverse to the direction of gravity and taking up the fine streams extruded from the extrusion surface against gravity (this process is referred to herein as an "upward spinning").

The spinneret used in the process of this invention is a mesh spinneret having many closely arranged small openings defined by an open ratio (α) of at least 10% preferably 20 to 90%, and preferably a mesh spinneret having many small openings defined by partitioning members of small width having elevations and depressions on its polymer extruding surface, said small openings being such that the polymer melt extruded through one small opening of the spinneret can move toward and away from the polymer melt extruded from another small opening adjacent to said one opening or vice versa through depressions of the partitioning members.

Since the spinneret used in this invention has many closely arranged small openings, the polymer melts extruded from adjacent small openings can move toward and away from each other. In particular, when the partitioning members defining the adjacent small openings have a depressed portion, the polymer melts can more readily move toward and away from each other through the depressed portion.

Desirably, the upward spinning process of this invention is carried out by turning the extrusion surface of the mesh spinneret upwardly such that the normal vector of the extrusion surface agrees completely with the direction of a vector ($-G$) which is quite reverse to the direction of gravity (G), or is different from it by only about several degrees.

The take-up direction of the fine streams extruded from the extrusion surface in the upward spinning may be the same as, or deviated by an angle of up to 30 degrees at most from, the normal vector direction of the extrusion surface.

According to the upward spinning process of this invention, the solidification length of the molten polymer can be made shorter than in the case of spinning it by using a spinneret whose extrusion surface is turned in the direction of gravity. The degree of the decrease of the solidification length differs depending upon the type of the polymer, the viscosity of the molten polymer, etc. Among polymers of the same type, the solidification length of a polymer having lower viscosity can generally be made shorter. It is easy to shorten the solidification length by not more than 10%.

Thus, according to the upward spinning process, the temperature of fine streams which have left the spinneret can be abruptly decreased over a shorter distance within a shorter period of time. It is easy therefore to produce as-spun fibers which can be easily converted into the elastic fibers of the invention by heat-treatment.

The aforesaid spinning process using a spinneret to be heated by Joule heat and the upward spinning process are described in detail in EP-A-47091.

In the process of this invention at least two different thermoplastic polymers are used. At least one of them has crystallinity capable of forming a row structure [this polymer is referred to hereinbelow as a

0 049 982

polymer (b)], and at least one other of them has crystallinity capable of forming lamellar crystals [this polymer is referred to hereinbelow as a polymer (c)].

Since the row structure is composed of lamellar crystals formed by folding of polymer molecule chains, the polymer (b) is embraced within the definition of the polymer (c).

5 Examples of preferred polymers (b) used in this invention include isotactic polypropylene, polybutene-1, poly(4-methylpentene-1), poly(3-methylbutene-1), poly(ϵ -capramide), poly(hexamethylene adipamide), polyoxymethylene, polyethylene sulfide, polytetramethylene terephthalate, polypivalolactone, and mixtures thereof.

10 Examples of preferred polymers (c) used in this invention include polyethylene, an ethylene/vinyl acetate copolymer, a polyester elastomer composed of a hard segment and a soft segment, polyethylene terephthalate ionomer, polyurethane, the polymers exemplified above as the polymer (b), and mixtures thereof.

The polymers (b) and (c) used in the invention preferably have such a high molecular weight as to be fiber-forming. For example, isotactic polypropylene has a melt index (MI) of preferably 0.1 to 30, more 15 preferably 0.5 to 20. Polybutene-1 has a melt index of preferably 0.1 to 30. Poly(4-methylpentene-1) has an MI of 1 to 70. Polyhexamethylene adipamide has an intrinsic viscosity, measured at 25°C in 99% formic acid, of preferably at least 0.8, more preferably 1 to 2.2. Poly(ϵ -capramide) has an intrinsic viscosity, measured at 25°C in m-cresol, of preferably at least 0.8, more preferably 1 to 2.2. Polyoxymethylene has an 20 intrinsic viscosity, measured at 25°C in p-chlorophenol, of preferably 1.3 to 2.2. Polytetramethylene terephthalate has an intrinsic viscosity, measured at 25°C in o-chlorophenol, of preferably at least 0.5, more preferably 0.7 to 2.0. Polypivalolactone has an intrinsic viscosity, measured at 25°C in a 6:4 (by weight) mixture of phenol and o-chlorophenol, of preferably at least 0.7, more preferably 1 to 2.0.

Polyethylene used as the polymer (c) has an MI of preferably 0.1 to 50. The ethylene/vinyl acetate copolymer preferably has an ethylene unit content of 60 to 95% by weight, and an MI of 1 to 400.

25 A preferred example of the polyester elastomer composed of a hard segment and a soft segment is an elastomer in which the hard segment is composed of an aromatic polyester derived from terephthalic acid as a main acid component and an aliphatic glycol having 2 to 6 carbon atoms as a main glycol component and the soft segment is composed of an aliphatic polyether or an aliphatic polyester having a second order transition point of not more than 0°C. The weight ratio of the hard segment to the soft segment in the 30 polyester elastomer is preferably from 70:10 to 30:90. The hard segment and the soft segment may each consist of a single or a plurality of components.

A preferred aromatic polyester forming the hard segment is the one derived from at least 50 mole% of terephthalic acid as a main acid component and an aliphatic glycol having 2 to 6 carbon atoms.

35 Examples of the acid component besides terephthalic acid are isophthalic acid, orthophthalic acid, methylterephthalic acid, methylisophthalic acid, and aliphatic dicarboxylic acids having 6 to 12 carbon atoms. Examples of the glycols having 2 to 6 carbon atoms are ethylene glycol, tetramethylene glycol, propylene glycol, and hexamethylene glycol.

The aromatic polyester (hard segment) may also contain such a component as 2,2-bis(hydroxyethoxyphenyl) methane, bisphenols, hydroxycaprylic acid and hydroxyethoxybenzoic acid.

40 Examples of such aromatic polyesters are polyethylene terephthalate, polypropylene terephthalate, polytetramethylene terephthalate and polyhexamethylene terephthalate.

Aliphatic polyethers or aliphatic polyesters having a second order transition point of not more than 0°C are preferred as the soft segment. The aliphatic polyethers preferably have a carbon/oxygen atom ratio of from 1.8:1 to 4.5:1, and the aliphatic polyesters are preferably polyesters derived from aliphatic 45 dicarboxylic acids having 4 to 12 carbon atoms and aliphatic diols having 2 to 12 carbon atoms, or polyesters composed of aliphatic hydroxycarboxylic acids having 4 to 12 carbon atoms.

Preferred polyester elastomers are polyester elastomers whose hard segment is a polyalkylene tere- or iso-phthalate having polytetramethylene glycol as a part of the glycol component, for example the one derived from polytetramethylene glycol (preferably having an average molecular weight of 1,000 to 3,000) 50 and at least one of C₂—C₄ alkylene glycol such as ethylene glycol, trimethylene glycol and tetramethylene glycol (preferably, tetramethylene glycol) as the glycol component and terephthalic acid or both terephthalic acid and isophthalic acid as the acid component. Of these, polyester elastomers containing a polyester derived from polytetramethylene glycol and tetramethylene glycol as the glycol component and terephthalic acid or both terephthalic acid and isophthalic acid as the acid component are especially 55 preferred.

The polyester elastomer especially preferably contains polytetramethylene glycol in a proportion of 5 to 70% by weight.

The aforesaid polyethylene terephthalate has an intrinsic viscosity, measured at 25°C in o-chlorophenol, of 0.5 to 1.2.

60 The aforesaid polyurethane is preferably the one obtained by polyaddition reaction of the aliphatic polyether or polyester described hereinabove as examples of the soft segment of the polyester elastomer and an aliphatic diol having 2 to 6 carbon atoms with a diisocyanate compound, for example, 4,4'-diphenylmethane diisocyanate or 4,4' dicyclohexylmethane diisocyanate.

The ionomer as the polymer (c) is suitably the one obtained by polymerizing an α -olefin and a 65 carboxyl-containing ethylenic monomer and crosslinking the resulting copolymer with a metal ion through

the carboxyl group. Preferably, it has a melt index of 0.5 to 20, and a melting point of 80 to 110°C. Especially preferred is a copolymer of ethylene and methacrylic acid which is crosslinked with a sodium ion or a zinc ion, the latter being most preferred.

5 Isotactic polypropylene, polyoxymethylene and polytetramethylene terephthalate are preferred among the polymers (b).

Copolymers composed of a polymer segment forming a crystalline phase and a polymer segment forming an amorphous phase, such as the polyester elastomers described above, are preferably used as the polymer (c).

10 Specific examples of the polymers (b) and (c) are polymers well known to those skilled in the art.

When a polymer exemplified as the polymer (b) is to be used as the polymer (c), the two polymers (b) and (c) should of course differ from each other, for example in the case of a combination of isotactic polypropylene and polyoxymethylene. Even when both of the polymers have crystallinity capable of forming a row structure as in this example, the elastic fibers of the invention in which at least one of the polymers is the polymer (b) can be obtained by selecting suitable crystallization conditions because having regard to the molding conditions, the polymers differ from each other substantially in the crystallizing conditions (such as the temperature, the time or the heating medium). For example, when as-spun fibers composed of 50 parts by weight of isotactic polypropylene and 50 parts by weight of polyoxymethylene are heat-treated at a temperature of about 150°C for about 1 minute in accordance with the process to be described hereinbelow, the former becomes the polymer (b) and the latter, the polymer (c).

20 A combination of polymers (b) and (c) whose difference in melting point is from 0 to 100°C, preferably from 0 to 80°C, is preferred because it shows a crystal arrangement suitable for production of the elastic fibers of the invention and therefore gives the elastic fibers of the invention easily by heat-treatment.

According to this invention, it is especially preferred to use isotactic polypropylene as the polymer (b) and a polyester elastomer in which the hard segment is derived from polytetramethylene glycol and tetramethylene glycol as the glycol component and terephthalic acid or both terephthalic acid and isophthalic acid as the acid component as the polymer (c); or polyoxymethylene as the polymer (b) and a polyester elastomer in which the hard segment is derived from polytetramethylene glycol and tetramethylene glycol as the glycol component and terephthalic acid or both terephthalic acid and isophthalic acid as the acid component as the polymer (c); or polytetramethylene terephthalate as the polymer (b) and a polyester elastomer in which the hard segment is derived from polytetramethylene glycol and tetramethylene glycol as the glycol component and terephthalic acid or both terephthalic acid and isophthalic acid as the acid component as the polymer (c).

According to the process of this invention, at least two thermoplastic polymer including the polymers (b) and (c) can give the elastic fibers of the present invention either as a blended mixture or as conjugate fibers.

35 According to the process of this invention, the as-spun fibers obtained from at least two thermoplastic polymers (b) and (c) by the above spinning method are then heat-treated to give the elastic fibers of the invention.

The heat-treatment is effected by various methods, for example hot air circulation; radiation heating by an infrared heater or a sheath heater; contacting with a hot drum; and heating by such media as hot water, vapor or silicone oils. These methods may be used singly or in combination.

40 The suitable heat-treating method can be selected depending upon the type of the polymer (b) of the as-spun fibers. The optimal method can be easily determined experimentally. For example, dry heat treatment is generally desirable. But when poly(*e*-caprolactam) is used as the polymer (b), wet heat treatment is desirable.

45 The heat-treatment temperature is desirably within the below-specified range depending upon the melting point of the polymer (b) constituting the as-spun fibers.

50 [Melting point (K) of the polymer (b)] \times (0.8 to 0.98)
preferably,

[Melting point (K) of the polymer (b)] \times (0.9 to 0.97)

When the temperature exceeds 0.98 times the melting point (absolute temperature) of the polymer (b), blocking tends to occur as a result of the heat treatment. On the other hand, if the temperature is below 0.8 times the melting point (absolute temperature) of the polymer (b), the heat-treated fibers do not have the desired elastic properties, or the heat-treating time required for giving the desired elastic properties tends to be prolonged.

For example, as-spun fibers composed of isotactic polypropylene and a polyester elastomer are heat-treated at a temperature determined depending upon the melting point (168°C=441K) of the isotactic polypropylene, which is therefore 80 to 159°C (353 to 432 K), preferably 124 to 155°C (397 to 428 K).

60 The heat-treating time depends upon the heat-treating temperature but in some cases, a period of not more than 1 second is even sufficient. Since as-spun fibers obtained by the spinning method utilizing a spinneret to be heated by Joule heat have such a crystalline structure as can easily give elastic fibers having a row structure by heat-treatment, the heat-treatment for a relatively short period of time gives the elastic fibers of the invention. Generally, the heat-treating time is from several seconds to 60 minutes. The

0 049 982

as-spun fibers during the heat-treatment may be under tension or without tension. Preferably, they are under some tension because elastic fibers having higher elastic recovery can be obtained within the same treating time.

5 The heat-treated fibers may be subjected to cold drawing to give elastic fibers having increased elastic properties and latent heat crimpability. The cold drawing treatment can be carried out at a draw ratio of up to 2, preferably 1.01 to 1.5.

A series of steps for producing an elastic fibrous assembly by the process of this invention will now be described specifically with reference to Figure 2 which schematically shows the apparatus for performing the process of the invention. It should be understood that for simplicity, those devices and component parts which do not greatly affect the manufacturing process are omitted in Figure 2.

10 Figure 2 shows an embodiment in which an elastic fibrous assembly is formed from a spinneret in a direction reverse to the direction of gravity.

In Figure 2, hoppers 1a and 1b for storing thermoplastic polymers are shown. The polymers (b) and (c) may be blended in chip form and stored in one of these hoppers from where the polymer blend may be fed 15 into a die 6 having a spinneret 7 through a feeder 2a or 2b, a melt-extruder 3a or 3b and a conduit 5a or 5b. Alternatively, the polymer (b) may be stocked in one hopper, for example 1a, and the polymer (c) in the other, for example 1b, and fed into the die in the same way as above.

The production of the fibers of the invention is described further with reference to Figure 2 with regard to the case of blending the polymers (b) and (c) in chip form and stocking the blend only in the hopper 1a.

20 Referring to Figure 2, thermoplastic polymers are stocked in the hopper 1a from where they are supplied to the extruder 3a by means of the feeder 2a. The blended polymers melted by the extruder are fed to the extrusion die 6 in a fixed quantity by a gear pump 4a through a conduit 5a.

The die 6 generally includes a heater (not shown) for maintaining the polymers in the molten state and heating it to the desired temperature. A spinneret 7 is provided on the top part of the die 6. The polymers 25 extruding surface of the spinneret 7 are turned in a direction reverse to the direction of gravity. An electric current can be supplied to the mesh construction of the extrusion surface of the spinneret 7 through copper plates 8. Specifically, this can be achieved by connecting the current taken from a power supply to both ends of the mesh spinneret while adjusting the voltage and current by means of a transformer 9 and a slidac 10.

30 The molten polymers extruded from the mesh spinneret and converted into fine streams are cooled by a cooling fluid (such as air) supplied to the extrusion surface of the spinneret or to its vicinity through a feed device 11, and solidified. The solidified fibrous assembly 13 is taken up by a take-up roller 12. The feed device 11 serves to supply the cooling fluid uniformly at a certain speed toward the extrusion surface of the mesh spinneret 7 and to its vicinity so that the molten polymers converted into fine streams may be rapidly 35 solidified. Suitably, the feed device 11 has a nozzle or slit. Preferably, the speed and direction of the cooling fluid are determined so that the solidification length [P(s)] becomes not more than 2 cm. The solidification length [P(s)] means the distance ranging from the extrusion surface of the molten polymers to a point at which they are solidified as fibers.

35 The resulting fibrous assembly 13 is taken up upwardly by the take-up roller 12, and sent to a heat treatment step. Figure 2 shows a heat-treatment device 14 having a heater. This is a mere example, and may be partly modified. Or another type of heat treatment means may be used. Other take-up rollers 15 (preferably cooled with water or the like) shown in Figure 2 are operated such that the speed of take-up of the fibrous assembly by the take-up rollers 15 is almost the same as that of the fibrous assembly which passes through the take-up roller 12. The fibrous assembly may also be cold-drawn by passing it through a 45 cooling zone provided between the take-up rollers 15 and the draw rollers 16, and this is generally preferred.

According to the process of this invention, therefore, the fibrous assembly in the form of an elongated strip can be formed upwardly, as shown in Figure 2. It can be directly sent to subsequent steps, such as a heat-treatment step, a crimping step, a cutting step (formation of short fibers), a fiber-opening step or a 50 web-forming step.

It will be readily understood from Figure 2 that large quantities of fibrous assemblies can be produced by an apparatus which is on the whole very compact and simple.

The fine streams of molten polymers from the spinneret can be taken up in accordance with the process of this invention so that the packing fraction (PF) defined by the following equation becomes 10^{-4} to 10^{-1} which is much higher than that (on the order of 10^{-5} at most) in a conventional melt-spinning process.

$$60 \quad PF = \frac{1}{Da}$$

wherein Da is an apparent draft ratio.

The packing fraction (PF) represents the sum of the cross-sectional areas of the entire fibers of the fibers assembly formed per unit area of the fiber-forming area of the spinneret, and constitutes a measure 65 of the density of fibers spun from the fiber-forming area, that is, the high-density spinning property.

0 049 982

The apparent draft ratio (D_a) is defined by the following equation.

$$D_a = V_L/V_o$$

wherein

5 V_L is the actual take-up speed of the fiber assembly (cm/min), and
 V_o is the average linear speed (cm/min) of the polymer melt in the extruding direction when the polymer melt is extruded so as to cover the entire extrusion surface of the fiber-forming area of the spinneret.

10 The elastic fibers of the invention composed of at least two thermoplastic polymers are spun preferably by a spinning method using the aforesaid mesh spinneret, and then heat-treating the resulting as-spun fibers. The elastic fibers of this invention include those composed of a blended mixture of the polymer (b) and the polymer (c) and those in the form of conjugate fibers. Which of these two forms is to be produced is determined by the dispersed state of the polymers (b) and (c) in the polymer melt fed into the mesh spinneret. For example, according to the method which comprises blending the polymers (b) and (c) in chip form and extruding the mixture through an extruder as shown in Figure 2, the polymers (b) and (c) are relatively well mixed in the polymer melt fed into the mesh spinneret, and there are formed elastic fibers in which at least one polymer is finely dispersed in the fiber cross section as can be clearly observed from the photo of Figure 7. Figure 7 is a photograph of a cross section of a conjugate fiber composed of a phase of a blended polymer mixture obtained by the polymer chip blending method and a phase of a single polymer. It is clearly seen from this photograph that the mixed state of the two polymers in the blended mixture differs greatly from the mixed state of the conjugate fibers.

15 Alternatively, similar elastic fibers can be produced by stocking the polymers (b) and (c) separately in the hoppers 1a and 1b shown in Figure 2 and mixing the two polymers in a screw extruder provided at a position immediately before the die 6 after passage through the conduits 5a and 5b.

20 Furthermore, composite elastic fibers of the invention the polymers (b) and (c) respectively form relatively large polymer phases in the cross section of the fibers as clearly observed from the photo of Figure 6 can be provided by preparing a polymer melt in which the polymers (b) and (c) form a relatively large block when they have reached the mesh spinneret (as clearly seen from the photo of Figure 5) by a method involving using the static mixer previously proposed by the present inventors in EP-A-46035.

25 Various static mixing units used normally in the mixing of molten polymers can be used either singly or in suitable combinations as the mixer for use in the present invention. Other examples of the mixer that can be used in this invention include a porous mixer obtained by closely aligning and laminating many porous corrugated plates in the longitudinal direction at certain intervals, a porous mixer made by closely aligning and laminating many wire meshes of a plain weave and/or twill weave in the longitudinal

30 direction, and a thin porous mixer made by closely filling and aligning many minute metallic balls and sintering them and thus bonding them to each other. Commercially available static mixers include, for example, a static mixer of Kenics Corp., a Sulzer static mixing unit of Gebrüder Sulzer AG, Ross ISG mixer of Charles Ross Co., a square mixer of Sakura Seisakusho, a Komax mixer of Komax System, Co., and a Bayer continuous mixer of Bayer AG.

35 Another static mixer which may be used in this invention is characterized by the fact that

- (a) it is a laminate made of a plurality of plates having a depressed portion,
- (b) the depressed portion of each of said plates forms a polymer melt inlet and a polymer melt outlet communicating with the polymer melt inlet,
- (c) said plates are comprised of at least two types of plates having differently-shaped depressed portions,
- (d) the polymer melt inlets of plates having depressed portions of the same shape form a common inlet for the same polymer melt, and thus the laminate has at least two common inlets for at least two different polymer melts, and
- (e) the polymer melt outlets are formed so as to give at least two different polymer melt flows adjoining each other.

40 In the present specification, the mixer having the above constituent elements (a) to (e) is referred to as a "laminated plate-type static mixer". As far as the present inventors know, such a laminate plate-type static mixer is a new type of mixer not known heretofore. By using this type of mixer, there can be easily obtained a molten macroblend in which a number of molten phases of at least two dissimilar polymers are coalesced in a lamellar structure, i.e. in a thin laminar flow. According to the laminated plate-type static mixer, a very thin layer-like melt can be obtained.

45 The depth (t_2) of the depressed portion is smaller than the thickness (t_1) of the plate, and it preferably satisfies the following expression.

50
$$0.2 t_1 \leq t_2 \leq 0.8 t_1$$

55 wherein t_1 is the thickness (mm) of the plate, and t_2 is the depth (mm) of the depressed portion.
 The especially preferred depth of the depression satisfies the following expression.

58
$$0.3 t_1 \leq t_2 \leq 0.7 t_1$$

60 wherein t_1 and t_2 are as defined hereinabove.

0 049 982

It is not necessary that all of the plates have the same thickness. Generally, however, plates of the same type desirably have the same thickness in order to obtain a homogeneous molten macroblend. It is especially advantageous to laminate at least two types of plates having the same thickness and the same depth in building the static mixer.

5 The thickness (t_1) of the plate is generally in the range of 0.05 to 2 mm, preferably 0.1 to 1 mm, especially preferably 0.2 to 0.7 mm.

The plate having a depressed portion has a width (W) of generally 5 mm to 10 cm, preferably 1 cm to 50 cm, and a height (H) of 5 mm to 50 cm, preferably 1 cm to 30 cm.

10 The static mixer may be provided within the die 6 and/or within the conduit 5a or 5b on the way from the conduit 5a or 5b to the die 6.

Figure 4 is a schematic vertical sectional view of one example of the die used in the process of this invention. It should be understood that Figure 4 shows the cross section of the die 6 shown in Figure 2 which is taken by cutting the mesh spinneret held by copper plates at both ends, nearly at its center at right angles (vertically) when viewed from above.

15 In Figure 4, the reference numeral 11 represents the die itself; 11, and 12, a flow passage of the molten polymers fed through the extruder 3, a gear pump 4 and the conduit 5 of Figure 2. The die 11 includes electric heaters 13-a and 13-b for maintaining the molten polymers at the desired temperature. The molten polymers which have been sent through the flow passage 12 are introduced into a reservoir 14 having a Kenics-type static mixer 17, and then rises upwardly slowly and stably while being mixed by the mixer 17.

20 Above the die 11 is installed a spinneret which is a mesh spinneret 15 in Figure 4. An area within which the molten polymer is extruded through small openings of the mesh spinneret and formed into a fibrous assembly has a width x. The mesh spinneret is firmly secured to the die 11 by means of fastening devices 16-a and 16-b. At those parts of the mesh spinneret which are held by the fastening devices, the openings of the mesh are blocked up with an inorganic adhesive, a high-melting or thermosetting resin, etc. to prevent flowing of electric current. In Figure 4, the direction of arrow means the direction reverse to the direction of gravity, and y=0 represents the position of the polymer extruding surface.

Cords are connected to copper plates attached to both ends (not shown) of the mesh spinneret 15 so as for permit flowing of an electric current.

In order to produce the conjugate elastic fibers of the invention using the aforesaid static mixer and 30 mesh spinneret, a combined polymer melt (i.e. molten macroblend) having many effective continuous boundary lines between the molten phases of the two dissimilar thermoplastic polymers is spun by using a spinneret in which one-fourth of the length of a partitioning member which defines one small opening in the spinneret is shorter than each of the aforesaid boundary lines.

35 The fiber cross section of the conjugated elastic fibers of the invention so produced has an average number of blocks ($\bar{N}(B)$) of preferably 1.5 to 20, especially preferably 2 to 5. $\bar{N}(B)$ means the average of the number of independent blocks of thermoplastic polymer which is determined by observing the cross sections of 30 arbitrarily selected fibers at an arbitrary position.

The conjugate elastic fibers of the invention may be said to have properties not seen in the elastic fibers of the invention composed of a blended mixture of relatively finely dispersed two or more 40 thermoplastic polymers because they can develop elasticity by the effect of the crystalline structure and also by the conjugate structure.

45 The elasticity based on the conjugate structure can be imparted when the elastic fibers of the invention are being heat-treated, or by subjecting the heat-treated fibers to another treatment. The elasticity based on the conjugate structure of the fibers can be also suitably imparted in the form of crimped fibers. Specifically the crimped fibers can be produced by heat-treating the conjugate elastic fibers under tension or under no tension in dry heat, boiling water, etc., or by simply drawing the conjugate fibers. The crimped fibers preferably have 1.2 to 7.9, especially 2.0 to 5.9 crimps per cm (3 to 20 crimps, especially 5 to 15 crimps, per inch).

50 Elastic fibers

The process of the invention provide elastic fibers having improved elastic properties, characterized by the fact that

(a) said elastic fibers are composed of at least two thermoplastic polymers,

(b) at least one of said thermoplastic polymers has crystallinity capable of forming a row structure,

55 (c) at least one other of said thermoplastic polymers has crystallinity capable of forming lamellar crystals, and

(d) said elastic fibers have a row structure, an elastic recovery ratio (ER) after 50% stretch of at least 70% and an elongation (EL) of from 200 to 700%.

60 The elastic fibers of the invention are novel in that they are composed of at least two thermoplastic polymers, and have a row structure in which lamellar crystals are stacked. It has been found in accordance with this invention that these elastic fibers can be produced by using at least one thermoplastic polymer capable of forming a row structure and at least one thermoplastic polymer capable of forming lamellar crystals.

65 The elastic fibers of this invention may contain the polymers (b) and (c) in any ratios. Preferably, these fibers contain the polymer (b) in an amount of at least 10% based on the entire polymers.

0 049 982

The elastic fibers of this invention have improved elastic properties over elastic fibers having a row structure and composed only of the polymer (b), fibers composed only of the polymer (c) [the polymer (c) includes a polymer which gives elastic fibers with the formation of a row structure, a polymer which gives elastic fibers even without forming a row structure, and a polymer which does not form a row structure nor give elastic fibers].

Generally, the elastic fibers, when containing a relatively large proportion of the polymer (b), tend to show higher elongation and lower initial modulus of elasticity than the elastic fibers composed only of the polymer (b) while showing much the same elastic recovery as the latter. Furthermore, the elastic fibers of the invention generally tend to show higher elastic recovery, elongation and initial modulus of elasticity than the fibers composed only of the polymer (c) when they contain a relatively large proportion of the polymer (c).

It will be clearly understood from specific working examples of the invention given hereinbelow that the elastic properties actually shown by the elastic fibers of the invention differ depending upon the types of the polymers (b) and (c), the state of mixing of the polymers (b) and (c), etc.

In order that the elastic fibers of the invention show improved elastic properties, especially those suitable for apparel applications, over the elastic fibers composed only of the polymer (b), those composed of a blended mixture of the polymers (b) and (c) contain the polymer (b) in a proportion of preferably at least 30% by weight, more preferably 45 to 95% by weight, based on the entire polymers, and those of conjugate type contain the polymer (b) in a proportion of at least 10% by weight, more preferably 25 to 90% by weight, above all 30 to 50% by weight, based on the entire polymers.

When the same polymers (b) and (c) are used, the blended mixture-type elastic fibers of the invention show a greater tendency to increased elongation and reduced initial modulus of elasticity than the conjugate-type elastic fibers of the invention at a relatively large polymer (b) proportion if the proportion of the polymer (c) is increased relative to the polymer (b).

Furthermore, when the same polymers (b) and (c) are used, the proportion of the polymer at which much the same elastic recovery as that of elastic fibers composed only of the polymer (b) is attained in a much wider range in the conjugate-type elastic fibers of the invention than in the blended mixture-type elastic fibers of the invention.

It has been found in accordance with this invention that the characteristic elastic properties of the blended mixture-type elastic fibers of the invention and the conjugated type elastic fibers of the invention can be exhibited by preparing at least one polymer phase of the conjugate-type fibers from a blended mixture consisting of a thermoplastic polymer (b) capable of forming a row structure and (c) a thermoplastic polymer capable of forming lamellar crystals.

The elastic fibers of the invention have an elastic recovery ratio (ER) after 50% stretch, as defined hereinbelow, of at least 70%, preferably at least 80%, an initial modulus of elasticity of preferably 0.45 to 13.5 g/dtex (0.5 to 15 g/de), more preferably 0.9 to 9.0 g/dtex (1 to 10 g/de), above all 1.3 to 4.5 g/dtex (1.5 to 5 g/de), an elongation of preferably 200 to 700%, more preferably 300 to 600%.

The elastic fibers of the invention further have a degree of orientation in the direction of the fiber axis of preferably at least 70%, more preferably at least 75%. In the row structure, the C axis of the crystals extends substantially in the direction of the fiber axis, and the crystals of the row structure have a long period of preferably 8 to 40 mm (80 to 400 Å), more preferably 10 to 40 nm (100 to about 400 Å).

By performing spinning on the mesh spinneret described hereinabove, the elastic fibers of this invention can be obtained as an assembly of many fibers. In addition to the aforesaid properties, these elastic fibers and assembly have the following properties.

When the fibrous assembly of the invention is cut at any position at right angles to the fiber axis, at least 90%, preferably at least 80%, especially preferably at least 70%, of the constituent fibers in this cross section have a non-circular cross-sectional shape. Most of the fibers constituting the assembly of the invention have a non-circular cross sectional shape.

According to this invention, the degree of cross-sectional non-circularity can be quantitatively expressed by the irregular shape factor (D/d) which is the ratio of the maximum distance (D) between two parallel lines circumscribing a fiber cross section to the minimum distance (d) between the two circumscribed parallel lines.

Each of the fibers having a non-circular cross section constituting the assembly of the invention preferably have an irregular shape factor of at least 1.1.

Furthermore, when the fibrous assembly of this invention is cut at an arbitrary position at right angles to the fiber axis, the cross sections of at least 50%, preferably at least 45%, especially preferably at least 40%, of the fibers differ from each other in at least one of shape and size.

According to this invention, the cross sections having a non-uniform shape and/or size can be distinguished by microscopic observations.

According to this invention, each of the fibers which constitute the fibrous assembly of the invention preferably has an irregular periodic variation in the size of cross sectional area along its longitudinal length.

The variation in the size of cross sectional area can be expressed by the intrafiber cross-sectional area variation coefficient [CV(F)] given by the following formula.

0 049 982

$$CV(F) = \frac{\sigma A}{\bar{A}}$$

5 Any 3 cm-length is selected in a given fiber of the fiber assembly, and the sizes of its cross-sectional areas taken at 1 mm intervals are measured by using a microscope. Then, the average (\bar{A}) of the sizes of the thirty cross-sectional areas, and the standard deviation (σA) of the thirty cross-sectional areas are calculated. Based on these values, $CV(F)$ can be computed in accordance with the above equation.

10 Each of the fibers which constitute the fibrous assembly of this invention preferably has an intrafiber cross-sectional area variation coefficient [$CV(F)$] in the range of 0.05 to 1.0.

According to the invention, the cross sections having different sizes can be determined quantitatively by the intra-assembly fiber cross-sectional area variation coefficient [$CV(A)$] given by the following equation

15

$$CV(A) = \frac{\sigma A}{\bar{S}(A)}$$

20 wherein $\bar{S}(A)$ is the average of the cross-sectional sizes of 100 fibers which are obtained by sampling at random a partial assembly of 100 fibers from the fibrous assembly of the invention, and microscopically measuring the cross-sectional sizes of the individual fibers in a cross section taken at an arbitrary position of the partial assembly, and σA is the standard deviation of the cross-sectional areas of the 100 fibers.

Fibers having different cross-sectional sizes which constitute the fibrous assembly of the invention have a $CV(A)$ of preferably 0.05 to 1.5, more preferably 0.1 to 1.5, especially preferably 0.2 to 1.

25 When a partial assembly of 100 fibers is sampled at random from the fibrous assembly of the invention and the cross sections of the individual fibers taken at an arbitrary position are observed microscopically, at least 50% of two cross sections sampled at random from the aforesaid cross sections preferably have

1) a shape distribution expressed by an irregular shape factor deviation ratio (α) of the following formula

30

$$\alpha = \frac{(D/d)_l - (D/d)_s}{(D/d)_l} \times 100 (\%)$$

35 wherein $(D/d)_l$ represents a larger irregular shape factor, and $(D/d)_s$ represents a smaller irregular shape factor, or

2) a size distribution expressed by a cross-sectional deviation ratio of the following formula

40

$$\beta = \frac{S_l - S_s}{S_l} \times 100 (\%)$$

wherein S_l is a larger cross-sectional size (mm^2), S_s is a smaller cross-sectional size (mm^2), and β is the cross-sectional area deviation ratio.

45 More preferably, the assembly of fibers in accordance with this invention is such that at least 50% of two cross sections sampled at random from the cross sections of the aforesaid fibers viewed by a microscope have

1) a difference in shape expressed by an irregular shape factor deviation ratio (α) of at least 2%, and/or

2) a difference in cross sectional area expressed by a cross-sectional area deviation ratio (β) of at least

50 5%.

It is desirable that when the fibrous assembly of the composite fibers of the invention is cut at an arbitrary position at right angles to the fiber axis, at least 50%, preferably at least 45%, more preferably at least 40%, of the fibers each have in their cross section at least two side-by-side coalesced blocks of at least two dissimilar thermoplastic polymer phases with at least a part thereof being exposed to the peripheral surface of the fiber, and at least one of the number, shape and size of the blocks vary from fiber to fiber. It should be understood that the side-by-side coalesced blocks exclude those blocks which are completely embraced within the fiber cross sections and are not exposed to the peripheral surfaces of the fibers.

At least 50% of the composite fibers which constitute the fibrous assembly of the invention have a cross section having at least two side-by-side coalesced blocks in accordance with the above definition. It will be readily appreciated from the description of the process of this invention that the ratio of cross sections having at least two side-by-side coalesced blocks can be varied depending upon the state of formation of a molten macroblend phase and the size of the small openings in the spinneret.

The number of side-by-side coalesced blocks should be construed to be the number of independent blocks at least a part of which is exposed to the peripheral surface of the fiber.

55 In the assembly of composite fibers in accordance with this invention, at least two blocks in each fiber

0 049 982

may respectively be composed of a single polymer phase, or of a microblend phase in which in a matrix of one polymer at least one other polymer is dispersed.

Thus, according to this invention, there is provided an assembly of fibers in which each block consists of a single polymer phase. There is also provided an assembly of fibers in which at least one block appearing in a fiber cross section by microscopic observation is coalesced with another block with a clear boundary line therebetween, said at least one block being composed of a matrix of at least one single polymer in which at least one other polymer is dispersed.

The fibrous assembly of fibers provided by this invention have an average fiber denier ($\bar{D}e$), as defined below, of 0.1 to 400 denier (0.111 to 444.4 dtex), preferably 0.3 to 200 denier 0.3333 to 222.2 dtex, more preferably 0.5 to 100 denier (0.5555 to 111.1 dtex).

The average denier size ($\bar{D}e$) in the assembly can be determined as follows:

Ten partial assemblies each consisting of 100 fibers are sampled at random from the fibrous assembly (for simplicity, three such partial assemblies may be used; the results are much the same for both cases), and each partial assembly is cut at one arbitrary position in the axial direction of fiber in a direction at right angles to the fiber axis. The cross section is then photographed through a microscope on a scale of about 2,000 times. The individual fiber cross sections are cut off from the resulting photograph, and their weights are measured. The total weight is divided by the total number of the cross-sectional microphotographs, and the result ($m(A)$) is calculated for denier (de).

Accordingly, the average denier size ($\bar{D}e$) in the assembly is calculated in accordance with the following equation

$$\bar{D}e = K \cdot m(A)$$

wherein $m(A)$ is the weight average value of the photographic fiber cross sections cut off; and K is a denier calculating factor defined by the equation

$$K = \frac{9 \times 10^5 \cdot \rho}{\gamma \cdot \delta}$$

in which γ is the weight (g) of the unit area of the photograph, δ is the ratio of area enlargement of the photograph, and ρ is the specific gravity of the thermoplastic polymers, all of these values being expressed in c.g.s. unit.

The elastic fibers of the invention may include heat stabilizers, nucleating agents, fire retarding agents, light stabilizers, slip agents, delusterants, pigments, plasticizers; thickeners, etc. which are usually employed as fiber additives.

The various properties described in the present application are measured by the following methods.

(1) Long periods of crystals based on the row structure

Using a small angle X-ray scattering device (Rotor Unit RU-3H, a product of Rigaku Denki Kabushiki Kaisha), a chart in the meridional direction is drawn within the range of 2θ =about 2° – 15° . The diffraction angle which shows a peak within this range is determined, and the long period is calculated by the Bragg's equation $2ds\sin\theta=\lambda$ (provided that $\lambda=0.15418$ nm (1.5418 Å) (CuKa)).

(2) Degree of orientation (DO) in the direction of the fiber axis

Using a broad angle X-ray scattering device (Rotor Unit RU-3H, a product of Rigaku Denki Kabushiki Kaisha), the position of a peak of a specified (h, k, l) plane reflection is determined from a chart in the equatorial direction, and a goniometer is fixed at that position. At this position, the goniometer is rotated through 180° (meridian—equator—meridian) within a plane perpendicular to the X-ray beams to draw a chart. The half value width (H, degree) of the chart is determined, and the degree of orientation is calculated from the following equation

$$\text{Degree of orientation (\%)} = \frac{180 - H}{180} \times 100$$

(3) Initial modulus of elasticity (IM)

From the stress-strain curve, a point of intersection between a tangent line at a point of 2% stretch and a tangent line at a point of 10% stretch is determined, and displacement (a %), stress (b g) and denier (1.111 dtex) (C denier) at the point of intersection are measured. The initial modulus is calculated from the following equation.

$$\text{Initial modulus (IM)} = \frac{b}{\frac{a}{100} \times C} \text{ (g/de)}$$

0 049 982

The stress-strain curve is prepared by cutting an 8 cm length of the fibers in the direction of the fiber axis, and pulling a 4 cm length sample taken from it by using a Tensilon (UTM-II-20, a product of Toyo Baldwin Co., Ltd) at a head speed of 100%/min.

5 (4) Elongation (EL)

This denotes the elongation at break which is read from the above stress-strain curve.

10 (5) Elastic recovery ratio (ER)

This means a stretch recovery ratio. A sample is stretched to 50% by using the aforesaid Tensilon at a head speed of 100%/min with a chuck distance of 4 cm (the length of the sample is 4 cm). Immediately then, the sample is returned to the original state at the same speed while releasing the stress. This cycle is repeated four times. The chuck distance (d cm) is measured at a time when the stress at the fifth pulling begins to increase from zero. The elastic recovery ratio is calculated from the following equation.

15

$$\text{Elastic recovery ratio (\%)} = \left(1 - \frac{d-4}{4}\right) \times 100$$

20 (6) Number of crimps (CN)

Sample monofilaments are cut to a length of 5.5 to 8.5.

A sample monofilament is cut to a length of 5.5 to 8.5, bonded to a paper frame while maintaining it loosened, and mounted on a crimp elasticity tester. The number of raised portions of the sample filament per 25 mm is calculated when an initial load of 1.8 mg/dtex (2 mg/de) is exerted.

The following Examples illustrate the present invention more specifically.

25

In these examples, all parts are by weight.

The polymers are sometimes expressed by the following abbreviations.

PP: isotactic polypropylene

Hytrel®: polyester elastomer

EVA: ethylene/vinyl acetate copolymer

30

Nylon 6: poly(ϵ -capramide)

PE: polyethylene

POM: polyoxymethylene

PBT: polytetramethylene terephthalate

PET: polyethylene terephthalate.

35

The properties given in the examples were measured with regard to a fibrous assembly having a denier size of about 333 dtex (300 de).

Example 1

40

Elastic fibers were produced by using an apparatus of the type shown in Figure 2.

Ninety parts of isotactic polypropylene (S115M, a product of Ube Industries, Ltd.) as the polymer (b)

and 10 parts of a polyester elastomer (Hytrel® 4056); melting point 168°C; a product of E. I. du Pont de Nemours & Co.) as the polymer (c) were fully mixed in chip form in a V-type blender, and dried at 140°C for 3 hours. The mixture was fed into hopper 1a. While being fed in a fixed amount by feeder 2a, the polymer

45

mixture was melted and kneaded in extruder 3a ($L/D=28$) having a screw diameter of 30 mm at a temperature of 230 to 290°C. The molten polymer was fed at a rate of 80 g/min by gear pump 4a to die 6

kept at 250°C, and discharged from a rectangular stainless steel 50-mesh plain weave wire mesh (a product of Nippon Kanaami K.K.) spinneret 7 having a length of 39 cm and a width of 2 cm and a molding area of 78 cm². A current of 30A (3V) was supplied to both ends of the spinneret through copper plates 8. That part of

50

the spinneret 7 which made contact with die 6 was insulated by coating an inorganic adhesive. A uniform cooling air was jetted against the molding area of the spinneret from cooling device 11 to form a solidified fibrous assembly 13. The fibrous assembly 13 was taken up by take-up roller 12 at a rate of 5 meters/min, heat-treated for 30 seconds through heat-treating box 14 held at a temperature of 150°C, and then cold-drawn to 1.5 times between rollers 15 and 16 through which cooling water was passed. Thus, an elastic fibrous assembly was obtained.

55 The manufacturing conditions and the various properties of the resulting fibers are shown in Table 1.

Examples 2 and 3

60

An elastic fibrous assembly was produced by the same procedure as in Example 1 except that the mixing ratio between isotactic polypropylene (S115M, a product of Ube Industries, Ltd.) and a polyester elastomer (Hytrel® 4056, a product of Du Pont) was changed. The manufacturing conditions and the properties of the resulting fibers are shown in Table 1.

Examples 4 to 6

65

Isotactic polypropylene (S115M) and an ethylene/vinyl acetate copolymer (Ultracene 625, a trademark

for a product of Toyo Soda Kogyo K.K.) were used as the polymer (b) and (c), respectively, and they were fully blended in chip form in a V-type blender and fed into hopper 1a. An electric current of 40A (4V) was supplied to the spinneret through copper plates 8. Otherwise, the same procedure as in Example 1 was repeated to form an elastic fibrous assembly.

5 The manufacturing conditions and the properties of the resulting fibers are shown in Table 1.

Examples 7 and 8

Isotactic polypropylene (S115M) as the polymer (b) and nylon 6 having an intrinsic viscosity, measured in m-cresol, of 1.34 as the polymer (c) were fully mixed in chip form by a V-type blender, dried at 140°C for 4 hours, and fed into hopper 1a. While being fed in a fixed quantity feeder 2a, the mixture was melted and kneaded by extruder 3a (L/D=28; screw diameter 30 mm) at a temperature of 270 to 300°C. Otherwise, the same procedure as in Example 1 was repeated to form an elastic fibrous assembly.

The manufacturing conditions and the properties of the resulting fibers are shown in Table 1.

15

Examples 9 and 10

Isotactic polypropylene (S115M) as the polymer (b) and polyethylene (J2522, a product of Ube Industries, Ltd.) as the polymer (c) were fully mixed in chip form in a V-type blender, and then fed into hopper 1a. Otherwise, the same procedure as in Example 1 was repeated to form an elastic fibrous assembly.

The manufacturing conditions and the properties of the resulting fibers are shown in Table 1.

Examples 11 to 13

Isotactic polypropylene (S115M) as the polymer (b) and a thermoplastic polyurethane elastomer (Roylar® E-85, a product of Uniroyal Company) as the polymer (c) were fully mixed in chip form in a V-type blender, dried at 110°C for 4 hours, and then fed into hopper 1a. A current of 50A (5V) was supplied to the spinneret. Otherwise, the same procedure as in Example 1 was repeated to form an elastic fibrous assembly.

Figure 8-a shows a broad angle X-ray photograph of isotactic polypropylene fibers heat-treated at 150°C for 30 seconds (Comparative Example 1) and Figure 8b is a broad angle X-ray photograph of elastic fibers of the invention obtained by spinning a mixture of isotactic polypropylene and 20% by weight of the aforesaid thermoplastic polyurethane and heat-treating the resulting fibers at 150°C for 30 seconds (Example 12). A comparison of Figure 8-a with Figure 8b shows that the orientation of the isotactic polypropylene crystals was promoted by blending the thermoplastic polyurethane with isotactic polypropylene.

The manufacturing conditions and the properties of the resulting fibers are shown in Table 1.

Examples 14 to 15

Isotactic polypropylene (S115M) as the polymer (b) and a polyester elastomer (Hytrel® 7246, a product of du Pont; melting point 218°C) as the polymer (c) were fully mixed in chip form in a V-type blender, dried at 150°C for 3 hours, and fed into hopper 1a. Otherwise, the same procedure as in Example 1 was repeated.

The manufacturing conditions and the properties of the resulting fibers are shown in Table 1.

Example 16

45 Elastic fibers were produced by using the same apparatus as shown in Figure 2.

Ninety parts of polyoxymethylene (Tenac® 5010, a product of Asahi Chemical Industry, Co., Ltd.) as the polymer (b) and 10 parts of a polyester elastomer (Hytrel® 4056) as the polymer (c) were fully mixed in chip form in a V-type blender, dried at 140°C for 3 hours, and then fed into hopper 1a. While being fed in a fixed quantity by feeder 2a, the mixture was melted and kneaded by extruder 3a (L/D=28; screw diameter 50 mm) at a temperature of 190 to 210°C. The molten mixture was fed into die 6 at 200°C at a rate of 80 g/min by gear pump 4a, and discharged from a 50-mesh stainless steel plain weave mesh spinneret 7 (made by Nippon Kanaami K.K.) having a rectangular molding area of 78 cm² (2 cm wide×39 cm long). A current of 70A (5V) was supplied to both ends of the spinneret through copper plates 8. A uniform cooling air was jetted against the molding area of the spinneret from cooling device 11 to give a solidified fibrous assembly 13, which was then taken up by take-up roller 12 at a rate of 5 m/min. The solidified fibrous assembly was heat-treated for 5 minutes in air at 150°C at constant length using a silicone rubber heater (a product of Sakaguchi Dennetsu K.K.), then cooled, and drawn to 1.5 times at room temperature to give an elastic fibrous assembly.

The manufacturing conditions and the properties of the resulting fibers are shown in Table 2.

60

Example 17

Seventy parts of the polyoxymethylene (Tenac® 5010) and 30 parts of the elastomer Hytrel® 4056 were fully mixed in chip form in a V-type blender, and then worked up by the same procedure as in Example 1 to form an elastic fibrous assembly. The manufacturing conditions and the properties of the resulting fibers are shown in Table 2.

Examples 18 and 19

5 Polyoxymethylene (Tenac® 5010) (dried at 150°C for 3 hours) as the polymer (b) and an ethylene-vinyl acetate copolymer (Ultracene 625) as the polymer (c) were fully mixed in chip form in a V-type blender, and fed into hopper 1a. Otherwise, the same procedure as in Example 16 was repeated to give an elastic fibrous assembly. The manufacturing conditions and the properties are shown in Table 2.

Examples 20 to 22

10 Isotactic polypropylene (S115M) and polyoxymethylene (Tenac® 5010) were fully mixed in chip form in a V-type blender, dried at 150°C for 3 hours, and then fed into hopper 1a. While being fed at a fixed quantity by feeder 2a, the mixture was melted and kneaded by extruder 3a (L/D=28; screw diameter 30 mm) at 190 to 210°C. The temperature of die 6 at this time was maintained at 200°C, and a current of 50A (4V) was supplied to the spinneret. Otherwise, the same procedure as in Example 1 was repeated to form an elastic fibrous assembly.

The manufacturing conditions and the properties of the resulting fibers are shown in Table 2.

Example 23

15 Fifty parts of isotactic polypropylene (S115M), 20 parts of polyoxymethylene (Tenac® 5010) and 30 parts of a polyester elastomer (Hytrel® 4056) were fully mixed in chip form in a V-type blender, dried at 140°C for 3 hours, and then fed into hopper 1a. Otherwise, the same procedure as in Examples 20 to 22 was 20 repeated to form an elastic fibrous assembly.

The manufacturing conditions and the properties of the resulting fibers are shown in Table 2.

Example 24

The same apparatus as shown in Figure 2 was used.

25 Ninety-five parts of polytetramethylene terephthalate having an intrinsic viscosity, measured in o-chlorophenol, of 1.0 as the polymer (b) and 5 parts of a polyester elastomer (Hytrel® 7246; melting point 218°C) as the polymer (c) were fully mixed in chip form in a V-type blender, dried at 140°C for 4 hours, and fed into hopper 1a. While being fed in a fixed quantity by feeder 2a, the mixture was melted and kneaded by extruder 3a (L/D=28; screw diameter 30 mm) at 260 to 300°C. The molten polymer was fed into die 6 at 30 280°C at a rate of 80 g/min by means of gear pump 4a, and discharged from 50-mesh stainless steel plain weave mesh spinneret 7 (a product of Nippon Kanaami K.K.) having a rectangular molding area of 78 cm² (2 cm wide×39 cm long). A current of 78A (7V) was supplied to both ends of the spinneret through copper plates 8. A uniform cooling air was jetted out against the molding area of the spinneret from cooling device 11 to give a solidified fibrous assembly 13. The fibrous assembly was then taken up by take-up roller 12 at a rate of 5 m/min, and heat-treated at constant length in a silicone oil (SH710, a product of Toray Silicone Co., Ltd.) at 200°C for 10 minutes, washed with n-hexane to remove the silicone oil, drawn to 1.3 times at room temperature to give an elastic fibrous assembly. The properties of the fibrous assembly are shown in Table 3.

35 Figure 9-a is an electron microphotograph (30,000×) of the surface of fibers obtained by etching the above fibers (before heat-treatment in the silicone oil) in a 70% aqueous solution of monoethylamine at 40 30°C for 30 hours; and Figure 9-b is an electron microphotograph (30,000×) of the surface of fibers obtained by etching the fibers (after heat-treatment in the silicone oil at 200°C) in the same way. The fibers before heat-treatment were seen to have a row structure-like crystals, and after the heat treatment, the crystals had a compact row structure.

Example 25 and 26

45 The same procedure as in Example 24 was repeated except that the mixing ratio between polytetramethylene terephthalate and the polyester elastomer was changed. The properties of the resulting elastic fibrous assembly are shown in Table 3.

Example 27 and 28

50 An elastic fibrous assembly was produced by the same procedure as in Example 24 except that polytetramethylene terephthalate having an intrinsic viscosity, measured in o-chlorophenol, of 1.0 and a polyester elastomer (Hytrel® 4056; m.p. 168°C) were used as the polymers (b) and (c), respectively. The properties of the resulting fibrous assembly are shown in Table 3.

Example 29

The same apparatus as shown in Figure 2 was used.

55 Isotactic polypropylene (S115M) as the polymer (b) was stocked in hopper 1a, and a polyester elastomer (Hytrel® 4056) was dried at 140°C for 3 hours and then stocked in hopper 1b. These polymers were fed at fixed quantities into extruders 3a and 3b having a screw diameter of 30 mm through feeders 2a and 2b respectively. In extruder 3a, the polymer was melted at 250 to 280°C, and in extruder 3b, the polymer was melted at 220 to 260°C. The molten polymers were fed in an amount of 70 parts at gear pump 4a and 30 parts at gear pump 4b and conducted to a section 5ab through sections 5a and 5b. The section 5ab contained a static mixer (4 elements), and the two polymer components were mixed there. The mixed

0 049 982

molten polymer was conducted to die 6 kept at a temperature of 250°C, further divided and mixed at a 4-element static mixer section, and discharged through a 50-mesh stainless steel plain weave mesh spinneret 7 (a product of Nippon Kanaami K.K.) having a rectangular molding area of 78 cm² (2 cm wide×39 cm long). The rate of discharge was 200 g/min.

5 The molten polymer just before extrusion from the mesh spinneret was sampled, and solidified. Its cross section was photographed and is shown in Figure 5. It is seen from Figure 5 that the polymers (b) and (c) were mixed as large blocks.

An electric current of 30A (3V) was supplied to both ends of the spinneret through copper plates 8. That part of the spinneret 7 which contacted the die 6 was insulated by coating an inorganic adhesive. A uniform cooling air was jetted against the molding area of the spinneret from cooling device 11 to give a solidified fibrous assembly. The fibrous assembly was taken up by take-up roller 12 at a rate of 10 m/min, and heat-treated for 30 seconds in heat-treating box 14 at 150°C. It was further cold-drawn to 1.5 times between rollers 15 and 16 through which cooling water was passed to give an elastic fibrous assembly. The properties of the fibrous assembly are shown in Table 4.

15 Figure 6 is a photograph of the cross section of the resulting fibrous assembly in which white parts show polypropylene and black parts, Hytrel® 4056. The photograph shows that the blocks were aligned side by side in a cross section of the fibers.

Examples 30 to 32

20 An elastic fibrous assembly was produced by the same procedure as in Example 1 except that the amounts of the isotactic polypropylene (S115M) and the polyester elastomer (Hytrel® 4056) were changed. The properties of the resulting fibers are shown in Table 4.

Examples 33 to 35

25 Isotactic polypropylene (S115M) was stocked in hopper 1a, and an ethylene/vinyl acetate copolymer (Ultracene 625) was stocked in hopper 1b. These polymers were melted respectively in extruder 3a kept at 240 to 280°C, and extruder 3b kept at 190 to 250°C. A current of 20A (2V) was supplied to the spinneret, and the ratio of the polymers fed was changed. Otherwise, the same procedure as in Example 29 was repeated to give an elastic fibrous assembly. The properties of the resulting fibrous assembly are shown in Table 5.

30 Examples 36 to 38

Isotactic polypropylene (S115M) was stocked in hopper 1a, and polyethylene terephthalate having an intrinsic viscosity, measured in o-chlorophenol, of 0.71 was dried at 150°C for 5 hours and then stocked in hopper 1b. These polymers were respectively melted in extruder 3a kept at 240 to 280°C, and extruder 3b kept at 270 to 300°C. The die 6 was kept at 270°C. A current of 70A (6.5V) was supplied to the spinneret. The ratio of the polymers fed was changed. Otherwise, the same procedure as in Example 29 was repeated to give an elastic fibrous assembly. The properties of the resulting fibrous assembly are shown in Table 5.

Examples 39 to 41

40 The same apparatus as shown in Figure 2 was used.

Polyoxymethylene (Tenac® 5010) was dried at 150°C for 3 hours and then stocked in hopper 1a, and a polyester elastomer (Hytrel® 4056) was dried at 140°C for 3 hours and stocked in hopper 1b. These polymers were respectively melted in extruder 3a kept at 190 to 210°C, and extruder 3b kept at 220 to 260°C. The die 6 having a 50-mesh plain weave mesh spinneret 7 was kept at 200°C. A current of 60A (5V) was supplied to the spinneret. The solidified fibrous assembly 13 was taken up at a rate of 10 m/min, and heat-treated at fixed length in air at 150°C for 5 minutes. After cooling, the fibrous assembly was cold-drawn to 1.5 times to give an elastic fibrous assembly. The properties of the resulting fibrous assembly are shown in Table 6.

50 Examples 42 and 43

The same apparatus as shown in Figure 2 was used.

Polytetramethylene terephthalate having an intrinsic viscosity, measured in o-chlorophenol, of 1.0 was dried at 150°C for 3 hours and stocked in hopper 1a, and a polyester elastomer (Hytrel® 7246) was dried at 150°C for 3 hours and stocked in hopper 1b. These polymers were respectively melted in extruder 3a kept at 260 to 290°C, and extruder 3b kept at 220 to 260°C. The die 6 having a 50-mesh plain weave mesh spinneret 7 was kept at 280°C. A current of 60A (5V) was supplied to the spinneret. The solidified fibrous assembly 13 was taken up at a take-up speed of 10 m/min, and heat-treated at fixed length in a silicone oil (SH710) at 200°C for 10 minutes. The heat-treated fibrous assembly was washed with n-hexane, and drawn to 1.5 times at room temperature to form an elastic fibrous assembly. The properties of the fibrous assembly are shown in Table 6.

Example 44

Sixty parts of isotactic polypropylene (S115M) and 40 parts of a polyester elastomer (Hytrel® 4056) were fully mixed in chip form in a V-type blender, dried at 140°C for 3 hours, and stocked in hopper 1a.

0 049 982

Isotactic polypropylene (S115M) was stocked in hopper 1b. Otherwise, the same procedure as in Example 29 was repeated to form an elastic fibrous assembly.

The cross section of the elastic fiber was photographed, and is shown in Figure 7. In Figure 7, a portion having fine deep and light shades shows the blended mixture from hopper 1a, and a white portion shows isotactic polypropylene.

5 The properties of the fibrous assembly are given in Table 7.

Examples 45 and 46

An elastic fibrous assembly was produced in the same way as in Example 44 except that the mixing ratio between the isotactic polypropylene (S115M) and the polyester elastomer (Hytrel® 4056) was varied. 10 The properties of the fibrous assembly are shown in Table 7.

Comparative Example 1

An elastic fibrous assembly was produced in the same way as in Example 1 except that only isotactic 15 polypropylene (S115M) was stocked in hopper 1a. The fibers obtained had a very high initial modulus of elasticity and a low elongation, and were unsuitable for practical application. The properties of the fibrous assembly are shown in Table 8.

Comparative Example 2

An elastic fibrous assembly was produced in the same way as in Example 16 except that 20 polyoxymethylene (Tenac® 5010) alone was dried at 15 140°C for 3 hours and stocked in hopper 1a. The resulting fibers had a high initial modulus of elasticity, and a low elongation. The properties of the fibrous assembly are shown in Table 8.

25 Comparative Example 3

An elastic fibrous assembly was produced in the same way as in Example 24 except that polytetramethylene terephthalate having an intrinsic viscosity, measured in o-chlorophenol, of 1.0 was dried at 150°C for 3 hours and stocked in hopper 1a. The resulting fibers had an elastic recovery ratio of as 30 low as 65% which is suggestive of an imperfect row structure. The properties of the fibrous assembly are shown in Table 8.

Examples 47 to 50

Polybutene-1 (a product of Scientific Polymer Products, Inc.), poly(4-methylpenten-1) (TPX® DX-245, a product of Mitsui Petrochemical Industries, Ltd.) or nylon 6 having an intrinsic viscosity of 1.34 was used as 35 the polymer (b). And an ethylene/vinyl acetate copolymer (Ultracene 625) or a polyester elastomer (Hytrel® 4056) was used as the polymer (c). These polymers were combined as shown in Table 9, and fully mixed in chip form in a V-type blender. The mixture was melted in an extruder having a screw diameter of 30 mm, and melt-spun to form a solidified fibrous assembly. The fibrous assembly was heat-treated to give an elastic fibrous assembly. The properties of the fibrous assembly are shown in Table 9.

40

45

50

55

60

65

TABLE 1

Example	1	2	3	4	5	6	7
Polymer (b)	PP	PP	PP	PP	PP	PP	PP
Parts by weight	90	70	50	90	70	50	90
Polymer (c)	Hytrel 4056	Hytrel 4056	Hytrel 4056	EVA	EVA	EVA	Nylon 6
Parts by weight	10	30	50	10	30	50	10
CV (A)	0.48	0.50	0.51	0.49	0.49	0.50	0.55
CV (F)	0.20	0.21	0.19	0.25	0.27	0.27	0.26
Cross-sectional shape	Tetragonal	ditto	ditto	ditto	ditto	ditto	ditto
Average monofilament dtex (de)	(6.5) 7.2	(7.0) 7.8	(7.0) 7.8	(6.5) 7.2	(7.0) 7.8	(6.5) 7.2	(7.0) 7.8
Irregular shape factor	1.2	1.25	1.26	1.2	1.26	1.30	1.25
CN	7.5	9.5	10.5	7	8.5	8.5	7
ER	92%	92	88	91	91	88	90
EL	350%	440	400	380	480	400	330
IM g/dtex (g/de)	(14.5) 13.1	(10.5) 9.5	(7.5) 6.8	(14) 12.6	(11) 9.9	(8) 7.2	(13.5) 12.2
DO	93.0%	93.3	92.8	93.0	93.0	92.5	93.0
Long period nm	17.0	17.0	16.8	17.1	17.0	16.7	16.9

0 049 982

TABLE 1 (continued)

Example	8	9	10	11	12	13	14	15
Polymer (b)	PP	PP	PP	PP	PP	PP	PP	PP
Parts by weight	70.	90	70	90	80	70	90	70
Polymer (c)	Nylon 6	PE	PE	Poly-urethane	Poly-urethane	Hytrel 7246	Hytrel 7246	
Parts by weight	30	10	30	10	20	30	10	30
CV (A)	0.54	0.51	0.50	0.58	0.59	0.59	0.49	0.48
CV (F)	0.25	0.25	0.25	0.30	0.30	0.32	0.20	0.19
Cross-sectional shape	ditto	ditto	ditto	ditto	ditto	ditto	ditto	ditto
Average monofilament dtex (dtx)	(7.0) 7.8	(7.0) 7.8	(6.5) 7.2	(6.5) 7.2	(7.0) 7.8	(6.5) 7.2	(7.0) 7.2	(7.0) 7.8
Irregular shape factor	1.25	1.25	1.25	1.35	1.35	1.35	1.2	1.2
CN	7	8.5	9.5	8.5	9	9.5	7	7.5
ER	88	92	90	93	92	90	92	91
EL	380	350	440	350	400	440	300	330
IM g/dtex (g/dtx)	(9.5) 8.4	(14) 12.6	(8.5) 7.7	(14.5) 13.1	(11.5) 10.4	(9.5) 8.4	(16.0) 13.5	(11.0) 9.9
DO	92.5	93.0	93.0	92.5	92.5	92.0	93.0	93.0
Long period nm	16.6	17.0	16.9	17.2	17.0	16.8	17.0	16.6

TABLE 2

Example		16	17	.18	19	20	21	22	23
Polymer (b)	POM	POM	POM	POM	P	P	P	P	
Parts by weight	90	70	90	70	90	70	60	60	
Polymer (c)	Hytex 4056	Hytrel 4056	EVA	EVA	POM	POM	POM	POM	
Parts by weight	10	30	10	30	10	30	50	Hytrel 4056	
CV (A)	0.48	0.52	0.53	0.51	0.53	0.50	0.49	0.50	
CV (F)	0.21	0.20	0.18	0.19	0.22	0.21	0.20	0.22	
Cross-sectional shape	Tetragonal	ditto	ditto	ditto	ditto	ditto	ditto	ditto	
Average monofilament dtex (d _e)	(8.0) 8.9	(8.5) 9.4	(8.0) 8.9	(8.5) 9.4	(8.5) 9.4	(8.0) 8.9	(8.0) 8.9	(7.5) 8.3	
Irregular shape factor	1.3	1.25	1.3	1.3	1.25	1.25	1.30	1.35	
CN	7.5	9.5	8.0	8.5	8.0	7.5	7.5	9.5	
Properties	ER	90%	89	90	89	91	90	89	
	EL	350%	350	350	400	430	420	440	
IM g/dtex (g/d _e)	(14.5) 13.1	(11) 9.9	(14.5) 13.1	(11) 9.9	(14.5) 13.1	(15.0) 13.5	(11.0) 9.9	(7.5) 6.8	
DO	80%	81	81	81.5	92	91.5	91.5	92	
Long period nm	14.5	14.4	14.2	14.2	17.0	17.3	16.8	16.8	

TABLE 3

Example		24	25	26	27	28
Polymer (b)	PBT	PBT	PBT	PBT	PBT	PBT
Parts by weight	95	90	80	95	90	
Polymer (c)	Hytrell 7246	Hytrell 7246	Hytrell 7246	Hytrell 4056	Hytrell 4056	
Parts by weight	6	10	20	5	10	
CV (A)	0.51	0.53	0.50	0.49	0.49	
CV (F)	0.20	0.19	0.18	0.18	0.19	
Cross-sectional shape	Tetragonal	ditto	ditto	ditto	ditto	
Average monofilament dtex (d _e)	(7.5) 8.3	(7.0) 7.8	(7.5) 8.3	(7.0) 7.8	(7.0) 7.8	
Irregular shape factor	1.2	1.25	1.25	1.2	1.25	
CN	7	8	8.5	6	6.5	
ER	85%	83	83	78	75	
EL	200%	200	230	200	200	
IM g/dtex (g/d _e)	(13.5) 12.2	(13.0) 11.7	(12.5) 11.3	(13.0) 11.7	(12.5) 11.3	
DO	91%	90	89	90	89	
Long period nm	14.4	14.0	14.0	13.6	13.5	

TABLE 4

Example	29	30	31	32
Polymer (b)	PP	PP	PP	PP
Parts by weight	70	50	30	10
Polymer (c)	Hytrel 4056	Hytrel 4056	Hytrel 4056	Hytrel 4056
Parts by weight	30	50	70	90
CV (A)	0.55	0.53	0.55	0.51
CV (F)	0.20	0.21	0.21	0.25
Cross-sectional shape	Tetragonal	ditto	ditto	ditto
Number of blocks	3.5	3.5	3.0	3.0
Form of blocks	side by side	ditto	ditto	ditto
Average monofilament dtex (dtx)	(7.5) 8.3	(7.5) 8.3	(7.0) 7.8	(7.0) 7.8
Irregular shape factor	1.3	1.3	1.4	1.45
CN	14	15	15	16
ER	92	92	92	92
EL	310	380	350	330
IM g/dtex (g/dtx)	(10) 9.0	(7) 6.3	(4) 3.6	(2) 1.8
DO	92.5%	92.5	92.5	92
Long period nm	17.0	16.8	16.8	16.5

TABLE 6

Example		33	34	35	36	37	38
Polymer (b)	PP	PP	PP	PP	PP	PP	PP
Parts by weight	70	50	30	90	70	70	60
Polymer (c)	EVA	EVA	EVA	PET	PET	PET	PET
Parts by weight	30	50	70	10	30	50	
Polymer combination							
CV (A)	0.51	0.55	0.53	0.49	0.51	0.52	
CV (F)	0.25	0.28	0.30	0.20	0.19	0.21	
Cross-sectional shape	Tetragonal	ditto	ditto	ditto	ditto	ditto	
Number of blocks	3.5	3.5	4.0	3.0	3.5	3.5	
Form of blocks	Side by side	ditto	ditto	ditto	ditto	ditto	
Average monofilament dtex (d _e)	(7.0) 7.8	(7.5) 8.3	(7.5) 8.3	(7.0) 7.8	(7.5) 8.3	(7.0) 7.8	
Irregular shape factor	1.4	1.45	1.45	1.3	1.3	1.3	
CN	11	12	12	13	13	13	
ER	92	92	92	92	91	91	
EL	350	330	330	430	400	350	
Properties							
IM g/dtex (g/d _e)	(10.5) 9.5	(7.5) 6.8	(4.5) 4.1	(13) 11.7	(11) 9.9	(9.5) 8.6	
DO	92.5%	93.0	93.0	92.0	92.0	92.0	
Long period nm	17.0	16.8	16.8	17.1	16.8	16.6	

TABLE 6

Example No.		39	40	41	42	43
Polymer (b)	POM	POM	POM	PBT	PBT	PBT
Parts by weight	70	50	30	90	80	
Polymer (c)	Hytrel 4056	Hytrel 4056	Hytrel 4056	Hytrel 7246	Hytrel 7246	
Parts by weight	30	50	70	10	20	
CV (A)	0.51	0.53	0.48	0.47	0.50	
CV (F)	0.19	0.20	0.19	0.21	0.21	
Cross-sectional shape	Tetragonal	ditto	ditto	ditto	ditto	
Number of blocks	4.0	3.5	3.5	4.0	4.5	
Form of blocks	Side by side	ditto	ditto	ditto	ditto	
Average monofilament dtex (d _e)	(8.5) 9.4	(8.0) 8.9	(8.0) 8.9	(7.5) 8.3	(8.0) 8.9	
Irregular shape factor	1.25	1.30	1.30	1.25	1.25	
CN	12	13	12	13	13	
ER	90%	89	89	72	76	
EL	300%	310	310	200	200	
IM g/dtex (g/d _e)	(11) 9.9	(7) 6.3	(5) 4.5	(11.5) 10.4	(11) 9.9	
DO	82	83	82	89	89	
Long period nm	15.0	14.8	14.6	13.3	13.4	

0 049 982

TABLE 7

Example			44	45	46
Polymer combination	Conjugate type	Polymer (b)	PP	PP	PP
		Parts by weight	42	35	36
		Polymer (c)	Hytrel 4056	Hytrel 4056	Hytrel 4056
		Parts by weight	28	35	54
		Polymer (b)	PP	PP	PP
		Parts by weight	30	30	10
		CV (A)	0.48	0.50	0.51
Properties of fibers	Conjugate type	CV (F)	0.21	0.20	0.20
		Cross-sectional shape	Tetragonal	ditto	ditto
		Number of blocks	3.5	4.0	3.5
		Form of blocks	side by side	ditto	ditto
		Average monofilament dtex (de)	(7.5) 8.3	(7.0) 7.8	(7.0) 7.8
		Irregular shape factor	1.2	1.2	1.3
		CN	13	14	13
Properties		ER	90%	90	89
		EL	450%	430	420
		IM g/dtex (g/de)	(10.5) 9.5	(10.5) 9.5	(8) 7.2
		DO	92.5%	92.8	92.6
		Long period nm	16.5	16.8	16.6

45

50

55

60

65

0 049 982

TABLE 8

Comparative Example		1	2	3
5	Polymer	PP	POM	PBT
Properties of fibers	CV (A)	0.46	0.54	0.48
	CV (F)	0.20	0.21	0.19
	Cross-sectional shape	Tetragonal	Tetragonal	Tetragonal
	Average monofilament dtex (de)	(6.5) 7.2	(8.5) 9.4	(7.5) 8.3
	Irregular shape factor	1.3	1.3	1.3
	CN	5	5.5	4.5
	ER	92%	90	65
	EL	200%	150	80
	IM g/dtex (g/de)	(20) 18.0	(20) 18.0	(15) 13.5
	DO	92%	80	89
Long period nm		17.0	14.4	13.0

TABLE 9

Example		47	48	49	50
35	Polymer combination	Polymer (b)	poly-butene-1	poly-butene-1	poly-(4-methyl-pentene-1)
40	Parts by weight	90	90	90	90
45	Polymer (c)	EVA	Hytrel 4056	Hytrel 4056	Hytrel 4056
Properties of fibers	Parts by weight	10	10	10	10
	CV (A)	0.52	0.55	0.52	0.50
	CV (F)	0.25	0.21	0.22	0.25
	Cross-sectional shape	Tetra-gonal	ditto	ditto	ditto
	Average monofilament dtex (de)	(7) 7.8	(7) 7.8	(7) 7.8	(7) 7.8
	Irregular shape factor	1.25	1.25	1.20	1.25
	CN	6.5	7.5	7.0	7.5
	ER	78	80	70	80
	IM g/dtex (g/de)	(11.5) 10.4	(10.0) 9.0	(12.5) 11.3	(14.5) 13.1

Claims

1. A fiber wherein
 - (a) said fiber is composed of at least two thermoplastic polymers,
 - (b) at least one of said thermoplastic polymers is capable of forming a row structure and
 - (c) at least one other of said thermoplastic polymers is capable of forming lamellar crystals, characterised in that
 - (d) said fiber is elastic, has a row structure, an elastic recovery ratio (ER) after 50% stretch of at least 70% and an elongation (EL) of from 200 to 700%.
- 10 2. An elastic fiber according to claim 1 wherein the C axis of the crystals of the row structure extends substantially in the direction of the fiber axis.
3. An elastic fiber according to claim 1 or 2 wherein the long period of crystals based on the row structure is from 8 to 40 nm (80 to 400 Å).
- 15 4. An elastic fiber according to any one of the preceding claims wherein crystals in the direction of the fiber axis have a degree of orientation of at least 70%.
5. An elastic fiber according to any one of the preceding claims which has an initial modulus of elasticity of from 0.45 to 13.5 g/dtex (0.5 to 15 g/de).
- 10 6. An elastic fiber according to any one of the preceding claims which is composed of a blended mixture of thermoplastic polymer (b) and thermoplastic polymer (c).
- 20 7. An elastic fiber according to any one of claims 1 to 5 which is a conjugate fiber composed of thermoplastic polymer (b) and thermoplastic polymer (c).
8. An elastic fiber according to claim 7 which is a conjugate fiber wherein at least one polymer phase is composed of a blended mixture of thermoplastic polymer (b) and thermoplastic polymer (c).
- 25 9. An elastic fiber according to any one of the preceding claims wherein the thermoplastic polymer (b) is isotactic polypropylene, polybutene-1, poly(4-methylpentene-1), poly(3-methylbutene-1), poly(ϵ -capramide), poly(hexa-methylene adipamide), polyoxymethylene, polyethylene sulfide, polytetramethylene terephthalate, polypivalolactone, or a mixture thereof.
- 10 10. An elastic fiber according to any one of the preceding claims wherein the thermoplastic polymer (c) is polyethylene, an ethylene/vinyl acetate copolymer, a polyester elastomer composed of a hard segment and a soft segment, polyethylene terephthalate ionomer, polyurethane, a polymer specified in claim 9 (except where the polymers (b) and (c) are the same), or a mixture thereof.
- 30 11. An elastic fiber according to any one of claims 1 to 8 wherein thermoplastic polymer (b) is isotactic polypropylene and thermoplastic polymer (c) is a polyester elastomer containing polytetramethylene glycol and tetramethylene glycol as the glycol component, and terephthalic acid or both terephthalic acid and isophthalic acid as the acid component.
- 35 12. An elastic fibrous assembly of a plurality of fibers as claimed in any one of the preceding claims.
13. A process for producing an elastic fibrous assembly as claimed in claim 12, which process comprises extruding a combined polymer melt of at least two thermoplastic polymers through a mesh spinneret, the combined polymer melt comprising at least 10% by weight of at least one thermoplastic polymer (b) which is capable of forming a row structure and not more than 90% by weight of at least one other thermoplastic polymer (c) which is capable of forming lamellar crystals, said spinneret including many closely arranged small openings and having an opening ratio (a), represented by the following formula, of at least 10%,

$$45 \quad a = \frac{V_a - V_t}{V_a} \times 100$$

in which V_a represents the total apparent volume of the spinneret which is taken within an unit area of its mesh portion, and V_t represents the total volume of partitioning members defining the small openings which is taken within a unit area of the mesh portion of the spinneret; said extrusion being carried out while generating Joule heat in the partitioning members of the spinneret and cooling the vicinity of the extrusion surface of the spinneret by supplying a cooling fluid, whereby the melt is converted into fine streams by the partitioning members; taking up and solidifying the fine streams without drawing them to provide as-spun fibers; and heat-treating the resulting as-spun fibers.

55 14. A process according to claim 13 wherein the extrusion surface of the spinneret is turned upward so that the normal vector of the extruding surface is reverse to the direction of gravity, and the fine streams extruded from the extrusion surface are taken up against gravity.

15. A process according to claim 13 or 14 wherein a combined polymer melt is used that has many effective continuous boundary lines between the molten phases of the two dissimilar thermoplastic polymers, each of which lines has a length greater than one-fourth of the length of a partitioning member which defines one small opening in the spinneret.

60 16. A process according to claim 15 wherein a combined polymer melt is used that has many effective continuous boundary lines between dissimilar molten polymer phases, each of which lines has a larger length than the length of each partitioning member which defines one small opening in the spinneret.

0 049 982

17. A process according to any one of claims 13 to 16 wherein after heat-treating, the fibrous assembly is cold-drawn at a draw ratio of not more than 2.

Patentansprüche

- 5 1. Eine Faser, bei der
 - (a) diese Faser aus mindestens zwei thermoplastischen Polymeren zusammengesetzt ist,
 - (b) mindestens eines dieser thermoplastischen Polymeren in der Lage ist, eine Reihenstruktur zu bilden und
- 10 (c) mindestens ein anderes dieser thermoplastischen Polymeren in der Lage ist, lamellare Kristalle zu bilden, dadurch gekennzeichnet, daß
 - (d) diese Faser elastisch ist, eine Reihenstruktur aufweist sowie ein elastisches Erholungsmaß (ER) von mindestens 70% nach Recken um 50% und eine Dehnung (EL) von 200 bis 700% besitzt.
- 15 2. Elastische Faser nach Anspruch 1, bei der die C-Achse der Kristalle der Reihenstruktur sich im wesentlichen in Richtung der Faserachse erstreckt.
- 20 3. Elastische Faser nach Anspruch 1 oder 2, bei der die lange Periode der Kristalle, bezogen auf die Reihenstruktur, 8 bis 40 nm (80 bis 400 Å) beträgt.
- 25 4. Elastische Faser nach einem der vorangehenden Ansprüche, bei der Kristalle in der Richtung der Faserachse einen Orientierungsgrad von mindestens 70% aufweisen.
- 30 5. Elastische Faser nach einem der vorangehenden Ansprüche, die einen Anfangs-Elastizitätsmodul von 0,45 bis 13,5 g/dtex (0,5 bis 15 g/de) aufweist.
- 35 6. Elastische Faser nach einem der vorangehenden Ansprüche, die aus einem mechanischen Gemisch aus thermoplastischem Polymeren (b) und thermoplastischem Polymeren (c) besteht.
- 40 7. Elastische Faser nach einem der Ansprüche 1 bis 5, die eine Konjugat-Faser ist, zusammengesetzt aus thermoplastischem Polymeren (b) und thermoplastischem Polymeren (c).
- 45 8. Elastische Faser nach Anspruch 7, die eine Konjugat-Faser ist, bei der mindestens eine Polymerphase aus einem mechanischen Gemisch eines thermoplastischen Polymeren (b) und eines thermoplastischen Polymeren (c) besteht.
- 50 9. Elastische Faser nach einem der vorangehenden Ansprüche, bei der das thermoplastische Polymer (b) isotaktisches Polypropylen, Polybuten-1, Poly(4-methylpenten-1), Poly(3-methylbuten-1), Poly(ε-capramid), Poly(hexamethylen-adipamid), Polyoxymethylen, Polyethylenulfid, Polytetramethylenterephthalat, Polypivalolacton oder ein Gemisch daraus ist.
- 55 10. Elastische Faser nach einem der vorangehenden Ansprüche, bei der das thermoplastische Polymer (c) Polyethylen, ein Ethylen/Vinylacetatcopolymer, ein Polyesterelastomer zusammengesetzt aus einem harten Segment und einem weichen Segment, Polyethylenterephthalat-ionomer, Polyurethan, ein Polymer der in Anspruch 9 angegebenen Art (ausgenommen, wenn die Polymeren (b) und (c) die gleichen sind) oder ein Gemisch hieraus ist.
- 60 11. Elastische Faser nach einem der Ansprüche 1 bis 8, bei der das thermoplastische Polymer (b) isotaktisches Polypropylen und das thermoplastische Polymer (c) ein Polyester-Elastomer ist, das Polytetramethylenglykol und Tetramethylenglykol als Glykolkomponente sowie Terephthalsäure oder sowohl Terephthalsäure als auch Isophthalsäure als Säurekomponente enthält.
- 65 12. Bündel aus mehreren elastischen Fasern, nach einem der vorangehenden Ansprüche.
- 70 13. Verfahren zur Herstellung eines Bündels aus einzelnen elastischen Fasern nach Anspruch 12, dadurch gekennzeichnet, daß man eine kombinierte Polymerschmelze aus mindestens zwei thermoplastischen Polymeren durch eine Netz-Spinndüse extrudiert, wobei die kombinierte Polymerschmelze mindestens 10 Gew.-% mindestens eines thermoplastischen Polymeren (b), das eine Reihenstruktur ausbilden kann, und nicht mehr als 90 Gew.-% mindestens eines anderen thermoplastischen Polymeren (c) enthält, das lamellare Kristalle ausbilden kann, wobei die Spinndüse zahlreiche nahe beieinander angeordnete kleine Öffnungen aufweist sowie ein Öffnungsverhältnis (a), wiedergegeben
- 75 50 durch die folgende Formel von mindestens 10%

$$a = \frac{V_a - V_t}{V_a} \times 100$$

- 55 in der V_a das gesamte scheinbare Volumen innerhalb einer Einheitsfläche seines Maschenbereiches und V_t das Gesamtvolume der unterteilenden Glieder, die die kleinen Öffnungen begrenzen, innerhalb einer Einheitsfläche des Maschenbereiches der Spinndüse bedeuten; wobei die Extrusion ausgeführt wird während Joule-Wärme in den unterteilenden Gliedern der Spinndüse erzeugt und die (unmittelbare) Nähe der Extrusionsfläche der Spinndüse gekühlt wird durch Zufuhr einer Kühlflüssigkeit, wodurch die Schmelze durch die unterteilenden Glieder in feine Ströme umgewandelt wird und daß man die feinen Ströme aufnimmt und verfestigt ohne sie zu ziehen, um wie-gesponnene Fasern zu erhalten, und (anschließend) die erhaltenen wie-gesponnenen Fasern wärmebehandelt.
- 60 14. Verfahren nach Anspruch 13, dadurch gekennzeichnet, daß die Extrusions-Oberfläche der Spinndüse nach oben gerichtet ist, so daß der senkrechte Vektor der Extrudier-Oberfläche entgegengesetzt

0 049 982

ist der Richtung der Schwerkraft und die feinen extrudierten Ströme von der Extrusions-Oberfläche gegen die Schwerkraft aufgenommen werden.

15. Verfahren nach Anspruch 13 oder 14, dadurch gekennzeichnet, daß eine kombinierte Polymerschmelze verwendet wird, die zahlreiche effektive kontinuierliche Grenzlinien zwischen den geschmolzenen Phasen der beiden unterschiedlichen thermoplastischen Polymeren aufweist, wobei jede dieser Linien länger ist als ein Viertel der Länge eines unterteilenden Gliedes, das eine kleine Öffnung in der Spinndüse begrenzt.

16. Verfahren nach Anspruch 15, dadurch gekennzeichnet, daß eine kombinierte Polymerschmelze verwendet wird, die zahlreiche effektive kontinuierliche Grenzlinien zwischen unterschiedlichen geschmolzenen Polymerphasen aufweist, wobei jede dieser Linien länger ist als die Länge jedes der unterteilenden Glieder, die eine kleine Öffnung in der Spinndüse begrenzen.

17. Verfahren nach einem der Ansprüche 13 bis 16, dadurch gekennzeichnet, daß nach der Wärmebehandlung das aus Einzelfasern bestehende Bündel bei einem Reckverhältnis von nicht mehr als 2 kalt gezogen wird.

15

Revendications

1. Fibre dans laquelle

20 (a) ladite fibre est composée d'au moins deux polymères thermoplastiques,
(b) au moins l'un desdits polymères thermoplastiques est capable de former une structure en alignement, et
(c) au moins un autre desdits polymères thermoplastiques est capable de former des cristaux lamellaires, caractérisée en ce que

25 (d) ladite fibre est élastique, possède une structure en alignement, un rapport de recouvrance élastique (RE) après un étirement de 50% d'au moins 70%, et un allongement (A) de 200 à 700%.

2. Fibre élastique conforme à la revendication 1, dans laquelle l'axe C des cristaux de la structure en alignement a sensiblement la direction de l'axe de la fibre.

3. Fibre élastique conforme à la revendication 1 ou 2, dans laquelle la grande période des cristaux, sur 30 la base de la structure en alignement, est de 8 à 40 nm (80 à 400 Å).

4. Fibre élastique conforme à l'une quelconque des précédentes revendications, dans laquelle les cristaux, dans la direction de l'axe de la fibre, ont un degré d'orientation d'au moins 70%.

5. Fibre élastique conforme à l'une quelconque des précédentes revendications, possédant un module initial d'élasticité de 0,45 à 13,5 g/dtex (0,5 à 15 g/de).

35 6. Fibre élastique conforme à l'une quelconque des revendications précédentes, composée d'un mélange intime de polymère thermoplastique (b) et de polymère thermoplastique (c).

7. Fibre élastique conforme à l'une quelconque des revendications 1 à 5, qui est une fibre conjuguée composée de polymère thermoplastique (b) et de polymère thermoplastique (c).

8. Fibre élastique conforme à la revendication 7, qui est une fibre conjuguée dans laquelle au moins 40 une phase polymère est composée d'un mélange intime de polymère thermoplastique (b) et de polymère thermoplastique (c).

9. Fibre élastique conforme à l'une quelconque des revendications précédentes, dans laquelle le polymère thermoplastique (b) est du polypropylène isotactique, du polybutène-1, du poly(4-méthylpentène-1), du poly(3-méthylbutène-1), du poly(ϵ -capramide), du poly(hexa-méthylène adipamide), du 45 polyoxyméthylène, du poly(éthylène-sulfure), du poly(tétraméthylène-téréphthalate), du polypivalolactone, ou un mélange de ceux-ci.

10. Fibre élastique conforme à l'une quelconque des précédentes revendications, dans laquelle le polymère thermoplastique (c) est du polyéthylène, un copolymère éthylène/(acétat de vinyle), un polyester élastomère composé d'un segment dur et d'un segment mou, un ionomère (polyéthylènetéréphthalate), du 50 polyuréthane, un polymère spécifié dans la revendication 9 (sauf si les polymères (b) et (c) sont identiques), ou un mélange de ceux-ci.

11. Fibre élastique conforme à l'une quelconque des revendications 1 à 8, dans laquelle le polymère thermoplastique (b) est du polypropylène isotactique, et le polymère thermoplastique (c) est un polyester élastomère contenant du poly(tétraméthyléneglycol) et du tétraméthyléneglycol comme composant glycol, 55 et de l'acide téréphthalique ou à la fois de l'acide téréphthalique et de l'acide isophthalique comme composant acide.

12. Corps fibreux élastique formé par assemblage de plusieurs fibres conformes à l'une quelconque des revendications précédentes.

13. Procédé de production d'un corps fibreux élastique conforme à la revendication 12, lequel procédé 60 comprend l'extrusion d'une masse fondue polymère combinée d'au moins deux polymères thermoplastiques à travers une filière à tamis, la masse fondue polymère combinée comprenant au moins 10% en poids d'au moins un polymère thermoplastique (b) capable de former une structure en alignement et pas plus de 90% en poids d'au moins un autre polymère thermoplastique (c) capable de former des cristaux lamellaires, ladite filière comprenant de nombreuses petites ouvertures disposées les unes près des autres, 65 et ayant un rapport d'ouverture (a), représenté par la formule suivante, d'au moins 10%,

0 049 982

$$\alpha = \frac{V_a - V_f}{V_a} \times 100$$

5 dans laquelle V_a représente le volume apparent total de la filière pris dans une surface unitaire de sa portion à tamis, et V_f représente le volume total des éléments de séparation définissant les petites ouvertures pris dans la surface unitaire de la portion à tamis de la filière; ladite extrusion étant effectuée tout en produisant de la chaleur par effet Joule dans les éléments de séparation de la filière et en refroidissant le voisinage de la surface d'extrusion de la filière en fournissant un fluide de refroidissement,
10 ce par quoi la masse fondue est transformée en des filets fins par les éléments de séparation; l'enlèvement et la solidification des filets fins sans les tirer afin de fournir des fibres comme filées; et le traitement par la chaleur des fibres comme filées résultantes.

14. Procédé conforme à la revendication 13, dans lequel la surface d'extrusion de la filière est tournée vers le haut de telle façon que le vecteur de la normale à la surface d'extrusion soit dirigé en sens inverse par rapport à la gravité, et que les filets fins extrudés de la surface d'extrusion soient enlevés à l'encontre de la gravité.

15. Procédé conforme à la revendication 13 ou 14, dans lequel on utilise une masse en fusion de polymères combinés qui présente de nombreuses lignes continues effectives de séparation entre les phases fondues des deux polymères thermoplastiques dissemblables, chacune de ces lignes possédant une longueur supérieure au quart de la longueur d'un élément de séparation qui définit une petite ouverture dans la filière.

16. Procédé conforme à la revendication 15, dans lequel on utilise une masse en fusion de polymères combinés qui présente de nombreuses lignes continues effectives de séparation entre les phases de polymère fondu dissemblables, chacune de ces lignes présentant une longueur supérieure à la longueur de chaque élément de séparation qui définit une petite ouverture dans la filière.

17. Procédé conforme à l'une quelconque des revendications 13 à 16, dans lequel, après le traitement à chaud, le corps fibreux est tiré à froid à un rapport de tirage qui n'est pas supérieur à 2.

30

35

40

45

50

55

60

65

0 049 982

Fig. 1-a

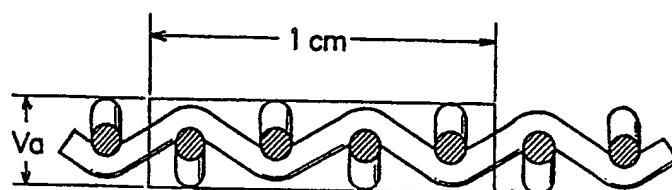
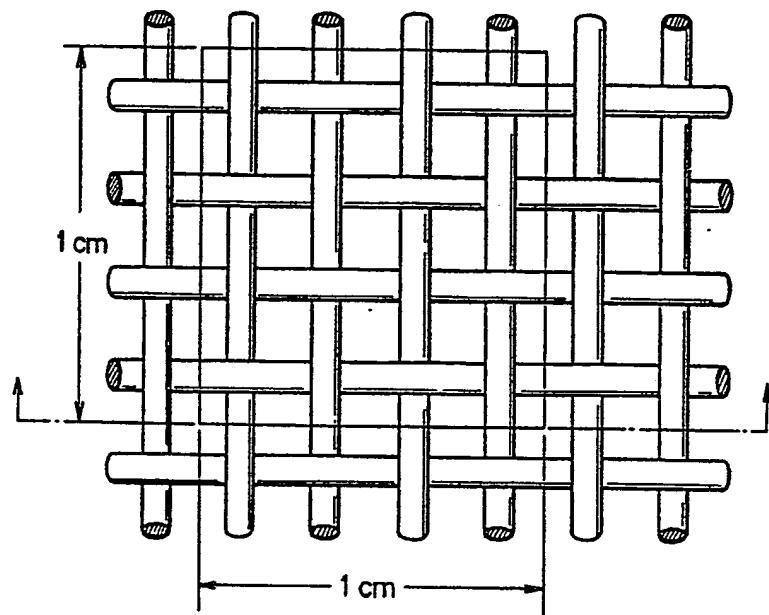
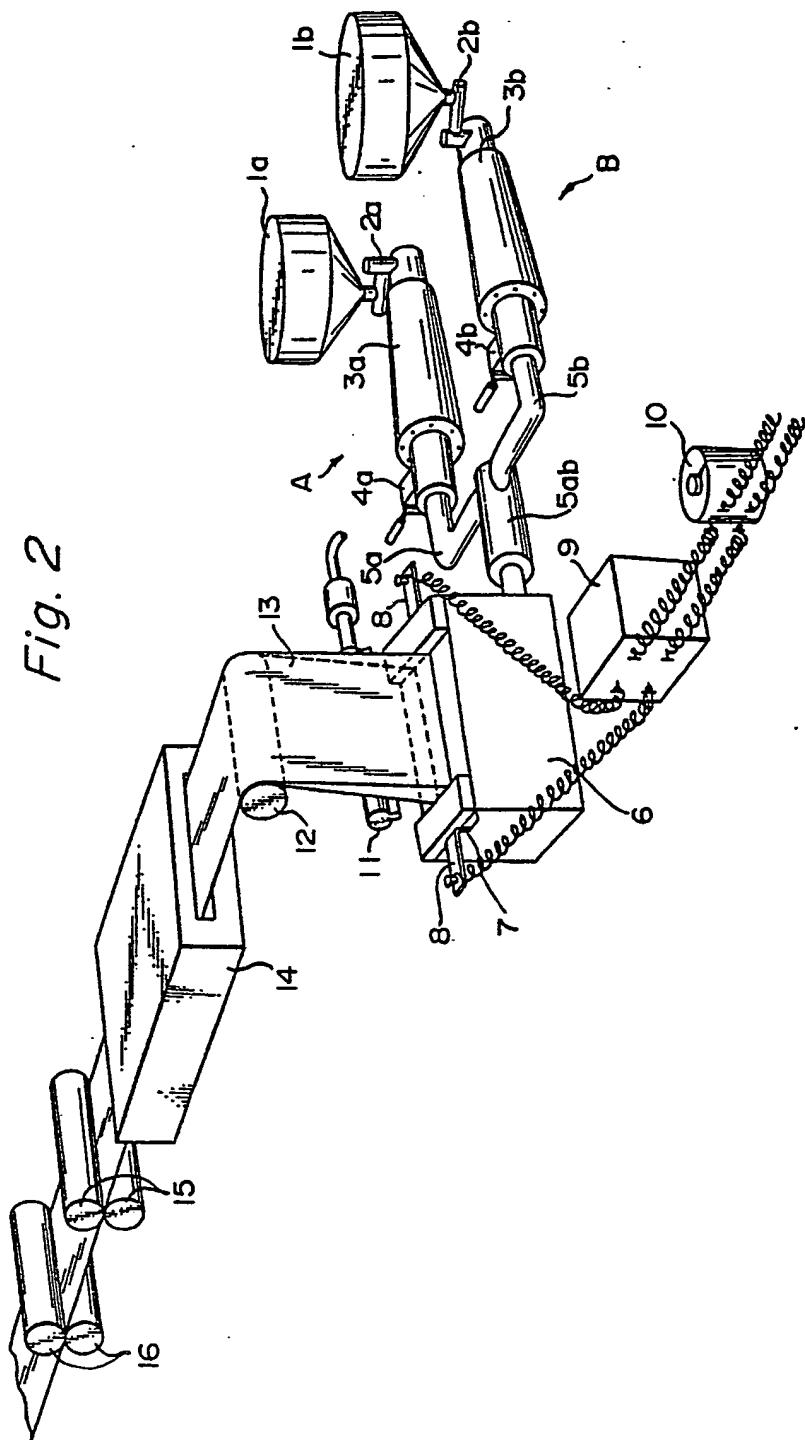
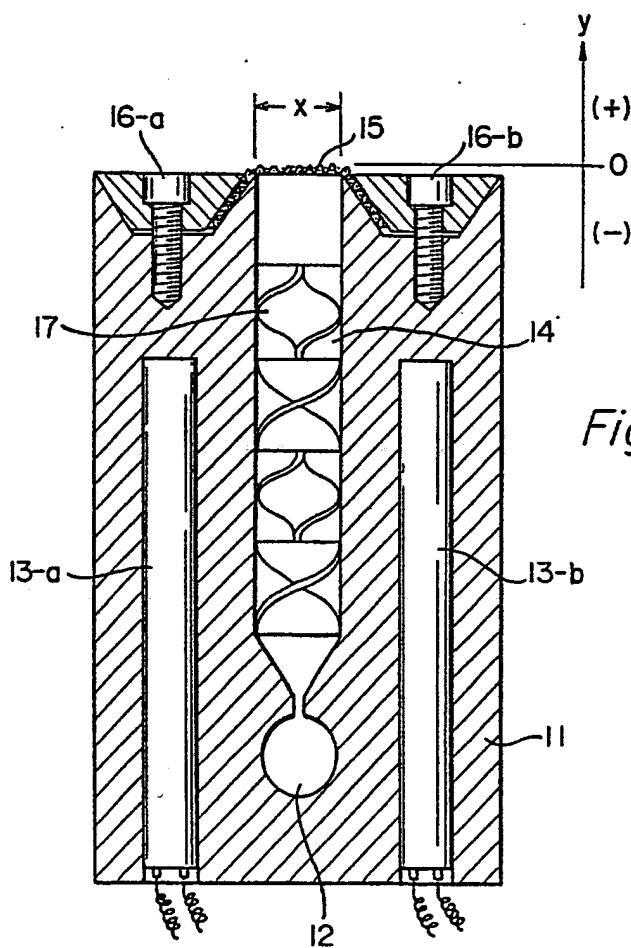
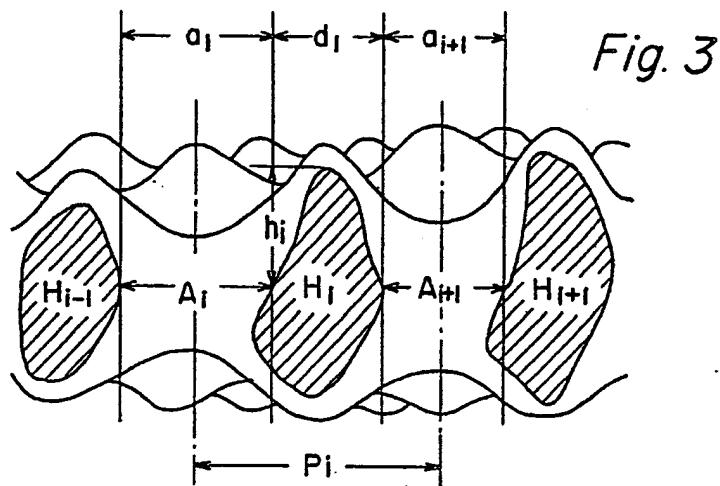


Fig. 1-b

0 049 982



0 049 982



0 049 982

Fig. 5

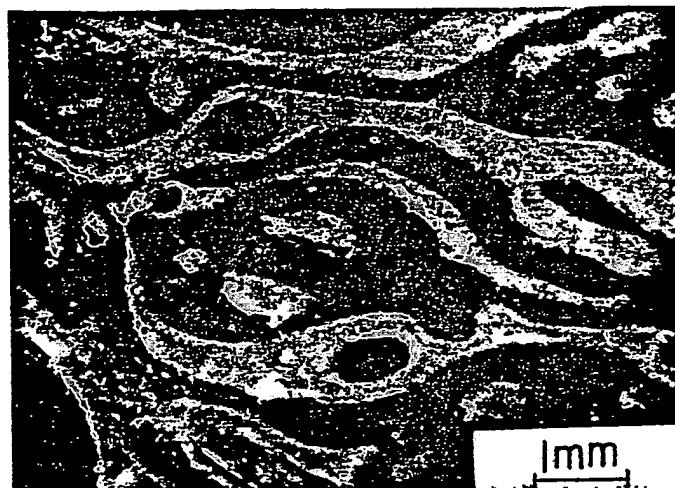


Fig. 6



0 049 982

Fig. 7

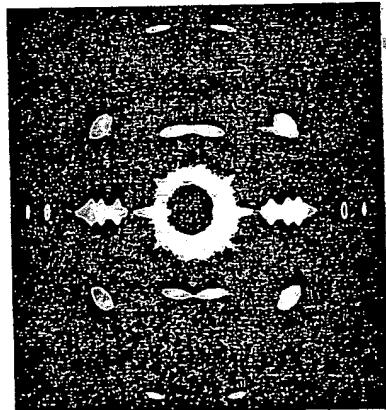


0 049 982

Fig. 8-a



Fig. 8-b



0 049 982

Fig. 9-a

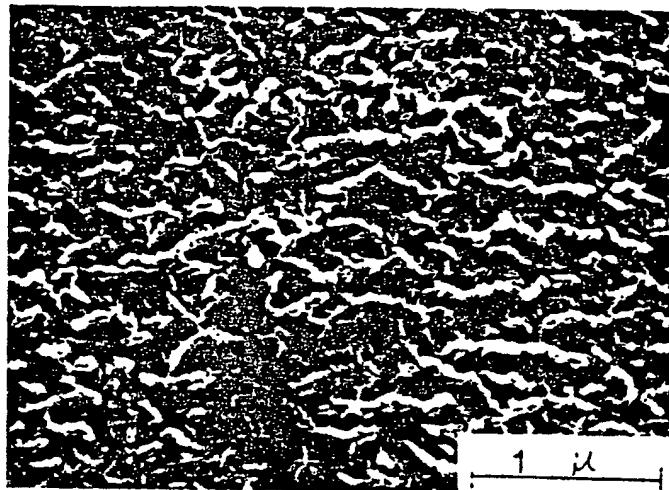
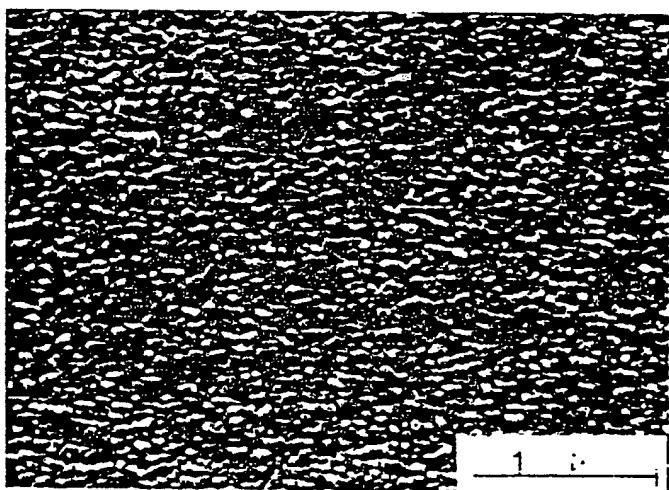


Fig. 9-b



**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- BLACK BORDERS**
- IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ~~EADED TEXT OR DRAWING~~**
- ~~BLURRED OR ILLEGIBLE TEXT OR DRAWING~~**
- SKEWED/SLANTED IMAGES**
- COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ~~GRAY SCALE DOCUMENTS~~**
- ~~LINES OR MARKS ON ORIGINAL DOCUMENT~~**
- REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- OTHER: _____**

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.